

5. POTENTIAL FOR HUMAN EXPOSURE

5.1 OVERVIEW

Aluminum, a silver-white, malleable, and ductile metal, is the most abundant metallic element in the lithosphere, comprising about 8% of the earth's crust. It is never found free in nature, but occurs combined with other elements, most commonly as aluminosilicates, oxides, and hydroxides in rock, minerals, clays, and soil. It is also present in air, water, and many foods. Bauxite, a weathered rock consisting primarily of aluminum hydroxide minerals, is the primary ore used in aluminum production. Aluminum enters environmental media naturally through the weathering of rocks and minerals. Anthropogenic releases are in the form of air emissions, waste water effluents, and solid waste primarily associated with industrial processes, such as aluminum production. Because of its prominence as a major constituent of the earth's crust, natural weathering processes far exceed the contribution of releases to air, water, and land associated with human activities.

The behavior of aluminum in the environment depends upon its coordination chemistry and the characteristics of the local environment, especially pH. The major features of the biogeochemical cycle of aluminum include leaching of aluminum from geochemical formations and soil particulates to aqueous environments, adsorption onto soil or sediment particulates, and wet and dry deposition from the air to land and surface water.

Aluminum is not bioaccumulated to a significant extent. Notable exceptions include some herbs and the tea plant, which can accumulate aluminum to 3,000-4,000 ppm and to 10,000 ppm respectively. Aluminum does not appear to accumulate to any significant degree in cow's milk or beef tissue and is, therefore, not expected to undergo biomagnification in terrestrial food chains. Similarly, because of its toxicity to many aquatic organisms, including fish, aluminum does not bioconcentrate in aquatic organisms to any significant degree. In order to bioaccumulate in the food chain, a substance cannot be acutely toxic to links in the chain; otherwise, the bioaccumulation stops.

Background levels of aluminum in rural air typically range from 0.005 to 0.18 ng/m³, whereas levels in urban and industrial areas can be considerably higher, ranging from 0.4 to 10 ng/m³. Concentrations of aluminum are highly variable in drinking water, ranging from <1 ppb to 1,029 ppb (Schenck et al. 1989). The use of alum (aluminum sulfate) as a flocculent in water treatment facilities is the usual cause of

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higher levels of aluminum in finished drinking water. The median level of aluminum in drinking water not receiving coagulation treatment is 0.043 mg/L, while that receiving coagulation treatment is 0.224 mg/L. Dissolved aluminum levels in surface and groundwater vary with pH and the humic acid content of the water. High aluminum concentrations in natural water occur only when the pH is < 5; therefore, levels in most surface water are very low. Aluminum is the third most common element in soil. Its concentration ranges widely from about 0.07% by weight (700 ppm) to 10% (100,000 ppm) with a typical concentration of about 7.1% (71,000 ppm).

Daily exposure to aluminum is inevitable due to its abundance and ubiquitous occurrence in nature and its diverse use by man. The general population is exposed to aluminum through consumption of food (including infant formula) and drinking water and through inhalation of airborne dust particulates, as well as through the use of such consumer items as antiperspirants, cosmetics, internal analgesics (buffered aspirins), anti-ulcerative medications, antidiarrheals, and antacids which contain aluminum compounds. The intake of aluminum from food and drinking water is very low, especially compared with that consumed by people taking aluminum-containing medicinal preparations, such as antacids. While aluminum is naturally present in food and water, the greatest contribution to aluminum in food and water by far is the aluminum-containing additives used in water treatment and processing certain types of food such as grain-based products and processed cheese.

Occupational exposures to aluminum occur during the mining and processing of aluminum ore into metal, recovery of scrap metal, production and use of aluminum compounds and products containing these compounds, and in aluminum welding. Individuals living in the vicinity of industrial emission sources and hazardous waste sites; individuals with chronic kidney failure requiring long-term dialysis or treatment with phosphate binders; patients requiring intravenous fluids; infants, especially premature infants fed soy-based formula containing high levels of aluminum; and individuals consuming large quantities of antacids, anti-ulcerative medications, buffered analgesics, antidiarrheal medications, or vitamins and food supplements may also be exposed to high levels of aluminum.

According to the Toxic Chemical Release Inventory, in 1996, total releases of aluminum to the environment (including air, water, and soil) from 264 large processing facilities were 5,605,000 pounds (TR196 1998). In addition, in 1996, total releases of aluminum oxide to the environment (including air, water, and soil) from 41 large processing facilities were 466,000 pounds (TR196 1998). Tables 5-1 and 5-2 list amounts released from these facilities grouped by state. The TRI data should be used with

Table 5-1. Releases to the Environment from Facilities that Manufacture or Process Aluminum and Its Compounds

State ^b	Number of facilities	Total of reported amounts released in pounds per year ^a						
		Air ^c	Water	Land	Underground injection	Total environment ^d	POTW transfer	Off-site waste transfer
AL	8	53,599	1,000	500	0	55,099	250	32,705
AR	10	77,326	5,800	255	0	83,381	5	57,868
AZ	1	255	0	0	0	255	0	250
CA	12	304,213	755	11,272	0	316,240	255	117,377
CT	1	380	0	0	0	380	0	920
GA	3	13,675	0	34,000	0	47,675	750	555,600
IA	7	3,242	250	17,820	0	21,312	250	312,809
IL	26	138,826	0	930,704	0	1,069,530	5	698,069
IN	24	195,904	20	30,250	0	226,174	264	2,592,215
KS	1	0	0	0	0	0	0	7,585
KY	10	285,965	516	1,218,950	0	1,505,431	371	509,755
LA	1	13	0	0	0	13	0	0
MA	3	260	0	0	0	260	0	930
MD	3	21,250	10	1,075	0	22,335	0	78,145
ME	1	12	0	0	0	12	0	13,580
MI	12	39,772	260	9,733	0	49,765	2,510	1,689,621
MN	5	17,637	6	0	0	17,643	0	293,302
MO	9	105,731	250	26,258	0	132,239	250	2,856,335
MS	1	24,750	0	750	0	25,500	6,500	11,000
NC	7	31,318	0	2,100	0	33,418	0	225,924
NJ	4	4,959	0	0	0	4,959	0	1,312
NY	4	1,522	0	0	0	1,522	0	56,000
OH	32	134,953	1,293	0	0	136,246	0	2,075,492

Table 5-1. Releases to the Environment from Facilities that Manufacture or Process Aluminum and Its Compounds (continued)

State ^b	Number of facilities	Total of reported amounts released in pounds per year ^a						
		Air ^c	Water	Land	Underground injection	Total environment ^d	POTW transfer	Off-site waste transfer
OK	4	7,857	0	0	0	7,857	0	597,816
OR	4	3,063	0	0	0	3,063	0	3,591,260
PA	22	36,102	35,778	633	0	72,513	250	829,067
SC	3	8,429	0	0	0	8,429	0	1,925,544
TN	16	33,378	68	1,587,770	0	1,621,216	6	6,397,157
TX	7	57,993	94	0	0	58,087	0	28,106
UT	3	11,635	0	0	0	11,635	0	45,400
VA	3	15,352	94	0	0	15,446	0	184,870
WA	2	482	0	0	0	482	0	260,002
WI	13	52,714	0	837	0	53,551	1,088	4,884,488
WV	2	700	2,795	0	0	3,495	0	5
Totals	264	1,683,267	48,989	3,872,907	0	5,605,163	12,754	30,930,509

Source: TRI96 1998

^aData in TRI are reported amounts released by each facility^bPost office state abbreviations used^cThe sum of fugitive and stack releases are included in releases to air by a given facility^dThe sum of all releases of the chemical to air, land, and water, and underground injection wells

POTW = publicly-owned treatment works

Table 5-2. Releases to the Environment from Facilities that Manufacture or Process Aluminum Oxide

State ^b	Number of facilities	Total of reported amounts released in pounds per year ^a						
		Air ^c	Water	Land	Underground injection	Total environment ^d	POTW ^e transfer	Off-site waste transfer
CA	2	692	0	0	0	692	0	98,279
CT	1	5	5	0	0	10	250	12,363
GA	2	750	250	45,000	0	46,000	0	64,300
IA	2	12,250	0	0	0	12,250	0	56,160
IL	1	0	0	0	0	0	250	41,200
IN	5	51,306	250	17,000	0	68,556	0	220,946
KY	2	5	0	0	0	5	323	11,133
MI	2	0	0	295,360	0	295,360	250	250
MN	1	255	0	0	0	255	0	0
MS	1	0	0	0	0	0	0	14,470
NC	1	0	0	0	0	0	0	64,980
NY	3	750	0	0	0	750	250	179,312
OH	7	24,214	0	0	0	24,214	0	414,294
PA	2	147	0	0	0	147	0	160,160
TN	2	0	0	0	0	0	5	56,036
TX	3	15,232	0	290	0	15,522	0	6,300
VA	1	0	0	0	0	0	0	36,000
WI	5	1,785	0	0	0	1,785	0	8,090,960
Totals	43	107,391	505	357,650	0	465,546	1,328	9,527,143

Source: TRI96 1998

^aData in TRI are reported amounts released by each facility^bPost office state abbreviations used^cThe sum of fugitive and stack releases are included in releases to air by a given facility^dThe sum of all releases of the chemical to air, land, and water, and underground injection wells^ePOTW = publicly-owned treatment works

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caution because only certain types of facilities are required to report (EPA 1995e). This is not an exhaustive list.

Aluminum has been identified in at least 427 of 1,428 hazardous wastes sites that have been proposed for inclusion on the EPA National Priorities List (NPL) (HazDat 1996). However, the number of sites evaluated for aluminum is not known. The frequency of these sites within the United States can be seen in Figure 5-1. Of these sites, 425 are located in the United States and 2 are located in the Commonwealth of Puerto Rico (not shown).

5.2 RELEASES TO THE ENVIRONMENT

Aluminum is released to the environment by both natural processes and anthropogenic sources. Because of its prominence as a major constituent of the earth's crust, natural processes far exceed the contribution of anthropogenic releases to the environmental distribution of aluminum (Lantzy and MacKenzie 1979). Anthropogenic releases are primarily to the atmosphere.

According to the Toxic Chemical Release Inventory, in 1996, a reported total of 5,605,000 pounds of aluminum were released to the environment (air, water, and soil) from 264 large processing facilities (TR196 1998). In addition, 466,000 pounds of aluminum oxide were released to the environment (air, water, soil) from 41 large processing facilities (TR196 1998). Tables 5-1 and 5-2 list amounts released from these facilities. An additional reported 12,754 pounds of aluminum were released by manufacturing and processing facilities to POTWs and a reported 30,931,000 pounds were transferred off-site (TR196 1998). A reported 1,328 pounds of aluminum oxide also were released by manufacturing and processing facilities to POTWs and an estimated 9,527,000 pounds of aluminum oxide were transferred off-site (TR196 1998). The TRI data should be used with caution because only certain types of facilities are required to report (EPA 1995e). This is not an exhaustive list.

5.2.1 Air

The largest source of airborne aluminum-containing particulates is the flux of dust from soil and the weathering of rocks (Lee and Von Lehmdehn 1973; Sorenson et al. 1974). In addition, a significant amount of aluminum-containing dust is generated by volcanic activity. Human activities, such as mining

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and agriculture, contribute to this wind-blown dust (Eisenreich 1980; Filipek et al. 1987). About 13% of atmospheric aluminum is attributed to anthropogenic emissions (Lantzy and MacKenzie 1979). The major anthropogenic sources of aluminum-containing particulate matter include coal combustion, aluminum production, and other industrial activities, such as smelting, that process crustal minerals (Lee and Von Lehmden 1973). Aluminum concentrations in air particulate emissions from iron and steel foundries and brass and bronze refineries range from about 100 to 1,000 ppm (Lee and Von Lehmden 1973). Que Hee et al. (1982) also found that aluminum was one of the most abundant elements quantified in coal stack emissions from power plants located in both the eastern and western United States. In addition, in U.S. cities, motor vehicle emissions contribute an estimated 0.9-9% of the observed elemental concentration of aluminum in these atmospheres (Ondov et al. 1982).

According to the Toxic Chemical Release Inventory, in 1996, the estimated releases of aluminum of 1,683,000 pounds to the air from 264 large processing facilities accounted for about 30% of total environmental releases (TR196 1998). Also, in 1996, the reported releases of aluminum oxide of 107,000 pounds to the air from 41 large processing facilities accounted for 23% of total environmental releases for this aluminum compound (TR196 1998). Tables 5-1 and 5-2 list amounts released from these facilities for aluminum and aluminum oxide respectively. The TRI data should be used with caution because only certain types of facilities are required to report (EPA 1995e). This is not an exhaustive list.

Aluminum has been identified in air samples collected at 9 of the 427 NPL hazardous waste sites where it has been detected in some environmental media (HazDat 1996).

5.2.2 Water

Aluminum occurs ubiquitously in natural waters as a result of the weathering of aluminum-containing rocks and minerals. Of the known geochemical responses to environmental acidification, the best documented is the mobilization of aluminum from terrestrial to aquatic environments (Campbell et al. 1992). This mobilization of aluminum is often episodic in nature and is associated with pH depressions (acidification) occurring during the spring snowmelt or associated with erosion from specific storm events (Campbell et al. 1992; Nelson and Campell 1991; Rosseland et al. 1990).

Aluminum levels in surface waters can be increased directly or indirectly by human activity through industrial and municipal discharges, surface run-off, tributary inflow, groundwater seepage, and wet and

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dry atmospheric deposition (Eisenreich 1980). For example, aluminum is released to surface waters in the effluent from bauxite processing and aluminum manufacturing facilities at concentrations that can be toxic to aquatic life (His et al. 1996; Trieff et al. 1995). However, the effluents of these facilities typically contain not only aluminum but a complex mixture of heavy metals such as iron, chromium and mercury, as well as minerals, silica, and other compounds, and synergistic effects of these metals and compounds cannot be ruled out. The use of aluminum sulfate and other aluminum compounds as coagulating agents in the treatment of raw drinking water supplies can significantly increase the total aluminum content in finished water (Malmberg 1985; Miller et al. 1984; Qureshi and Sung 1984). Weathering of sulfide ores exposed to the atmosphere in inactive mines and tailings dumps releases large quantities of sulfuric acid and metals such as aluminum (Filipek et al. 1987). Increasingly, acid environments caused by such acid mine drainage or by acid rain will subsequently cause an increase in the dissolved aluminum content of the surrounding waters (Brusewitz 1984; Filipek et al. 1987). In addition, atmospheric deposition is a source of aluminum input to surface water. The atmospheric loading of aluminum to Lake Michigan was estimated to be 5 million kg/year, of which 74% was to the southern basin where the influence of agricultural and industrial activity (e.g., steel manufacturing and cement production) was greatest (Eisenreich 1980).

According to the Toxic Chemical Release Inventory, in 1996, the reported releases of 48,989 pounds of aluminum to water from 264 large processing facilities accounted for 0.9% of the total environmental releases (TR196 1998). An additional 12,754 pounds of aluminum were released indirectly to POTWs and some of this mass ultimately may have been released to surface waters. Also, in 1996, the reported releases of 505 pounds of aluminum oxide to water from 43 large processing facilities accounted for 0.1% of the total environmental releases (TR196 1998). An additional 1,328 pounds were released indirectly to POTWs and some of this mass ultimately may have been released to surface waters. Tables 5-1 and 5-2 list amounts released from these facilities for aluminum and aluminum oxide, respectively. The TRI data should be used with caution because only certain types of facilities are required to report (EPA 1995e). This is not an exhaustive list.

Aluminum has been identified in surface water, leachate, and groundwater samples collected at 227,54, and 336 of the 427 NPL hazardous waste sites, respectively, where it has been detected in some environmental media (HazDat 1996).

5.2.3 Soil

Aluminum is the third most abundant element in the soil, constituting about 8% of the minerals (Rosseland et al. 1990). This element can be released naturally by the weathering of aluminum-containing rocks. Aluminum is also released to soil as a major constituent of many mining wastes and is also contained in solid wastes from coal combustion and aluminum reduction and other metal processing operations (Gabler and Stroll 1983; Krishnaswamy 1984).

According to the Toxic Chemical Release Inventory, in 1996, reported releases of 3873,000 pounds of aluminum to soil from 264 large processing facilities accounted for 69% of total environmental releases of aluminum (TR196 1998). Also, in 1996, reported releases of 358,000 pounds of aluminum oxide to soil from 43 large processing facilities accounted for 77% of total environmental releases (TR196 1998). No aluminum or aluminum oxide was released via underground injection in 1996. Tables 5-1 and 5-2 list amounts released from these facilities for aluminum and aluminum oxide, respectively. The TRI data should be used with caution because only certain types of facilities are required to report (EPA 1995e). This is not an exhaustive list.

Aluminum has been identified in soil and sediment samples collected at 203 and 151 of the 427 NPL hazardous waste sites, respectively, where it has been detected in some environmental media (HazDat 1996).

5.3 ENVIRONMENTAL FATE

5.3.1 Transport and Partitioning

Aluminum occurs widely in nature in silicates such as micas and feldspars, complexed with sodium and fluorine as cryolite, and in bauxite rock, which is composed of hydrous aluminum oxides, aluminum hydroxides, and impurities such as free silica (Cotton and Wilkinson 1988). Because of its reactivity, aluminum is not found as a free metal in nature (Bodek et al. 1988). Aluminum exhibits only one oxidation state (+3) in its compounds and its behavior in the environment is strongly influenced by its coordination chemistry. Aluminum partitions between solid and liquid phases by reacting and complexing with water molecules and anions such as chloride, fluoride, sulfate, nitrate, phosphate, and negatively charged functional groups on humic materials and clay.

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The transport and partitioning of aluminum in the environment is determined by the chemical properties of the element itself and the characteristics of the environmental matrix that affect solubility. At a pH >5.5, naturally occurring aluminum compounds exist predominantly in an undissolved form such as gibbsite, $\text{Al}(\text{OH})_3$, or as aluminosilicates except in the presence of high amounts of dissolved organic material or fulvic acid, which binds with aluminum and can cause increased dissolved aluminum concentrations in streams and lakes (Brusewitz 1984). Organic acids have been found to be important weathering agents for dissolving and transporting aluminum in an alpine soil environment (Litaor 1987). The ability of these organic acids to complex aluminum in sub-alpine soil solutions was found to increase as the pH rose from 3.8 to 5 (Dahlgren and Ugolini 1989). In this study, dissolved aluminum was found primarily as organic complexes when organic carbon/metal ratios were >50 (Dahlgren and Ugolini 1989).

In general, decreasing pH (acidification) results in an increase in mobility for monomeric forms of aluminum (Goenaga and Williams 1988), which is of concern with respect to the occurrence of acid rain and the release of acid mine drainage. Aluminum in soil solutions and surface waters in a mining region rich in metallic sulfides was in a labile form as Al-SO_4 and Al^{3+} species. Acidic conditions are created by the microbial oxidation of sulfides in tailing piles, resulting in sulfuric acid. In contrast, in areas not affected by acidification, aluminum in solution was partitioned between labile and non-labile forms, the latter being predominantly bound to fluorine (Alvarez et al. 1993). In soils, the most soluble form of aluminum under acidic conditions is nonsiliceous, organically-bound aluminum (Mulder et al. 1989).

In groundwater or surface water systems, an equilibrium with a solid phase or form is established that largely controls the extent of aluminum dissolution which can occur. In acid sulfate waters resulting from mine drainage, gibbsite and kaolinite are not stable, and the solubility of the minerals jurbanite ($\text{Al}(\text{SO}_4)(\text{OH})\cdot\text{H}_2\text{O}$) or alunite ($\text{KAl}_3(\text{SO}_4)_2(\text{OH})_6$) may control aluminum levels (Filipek et al. 1987). In a Colorado alpine watershed soil, the chemical equilibria of aluminum in interstitial water at a pH range of 4.4-7.2 were controlled by amorphous aluminosilicate rather than gibbsite (Litaor 1987).

In addition to the effect of pH on mobility, the type of acid entering environmental systems may also be important. Nitric acid was found to leach more aluminum from soil columns representative of high-elevation forest floor soils than did sulfuric acid (James and Riha 1989). This is most likely due to the higher solubility of aluminum nitrate than aluminum sulfate. However, in mineral horizons below the forest floor, the study found that concentrations of aluminum leached by these acids did not differ from concentrations of aluminum leached by distilled, deionized water at a pH of 5.7. The authors concluded

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that soluble constituents from the forest floor affected the aluminum solubility in the underlying mineral horizons under the leaching conditions that they used. These constituents may have included natural buffering agents which resist changes in pH and, therefore, negate or mediate the effect of the acid.

The ability of mineralized soil to control the migration of aluminum was observed in another study. Acidic leachate from coal waste containing aluminum was percolated through soil containing varying amounts of calcium carbonate (Wangen and Jones 1984). Soluble aluminum was found to decrease dramatically as the pH of the percolating leachate increased and aluminum oxide precipitates formed; at pH 6, no dissolved aluminum was measured. The authors concluded that alkalinized carbonaceous soils provide the best control material for acidic leachates from coal mineral wastes.

The adsorption of aluminum onto clay surfaces can be a significant factor in controlling aluminum mobility in the environment, and these adsorption reactions, measured in one study at pH 3.0-4.1, have been observed to be very rapid (Walker et al. 1988). However, clays may act either as a sink or a source for soluble aluminum depending on the degree of aluminum saturation on the clay surface (Walker et al. 1988).

The presence of high levels of suspended solids in stream surface water during storm episodes resulted in higher concentrations of adsorbed aluminum than in the absence of suspended solids (Goenaga and Williams 1988). The increased adsorption was not strictly linear, with higher concentrations of suspended solids due to variations in the particle size distribution and the nature of the particles.

Within the pH range of 5-6, aluminum complexes with phosphate and is removed from solution. Because phosphate is a necessary nutrient in ecological systems, this immobilization of both aluminum and phosphate may result in depleted nutrient states in surface water (Brusewitz 1984). Conversely, aluminum has been added to a nutrient-rich lake in Sweden with some success in an effort to arrest the "aging process" caused by an overabundance of phosphate (Jernelov 1971).

Aluminum salt coagulants are used in the treatment of potable drinking water, and unretained aluminum (approximately 11% of the added aluminum) was found to be transported through a water distribution system (Driscoll and Letterman 1988).

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Aluminum as a constituent of soil, weathered rock, and solid waste from industrial processes, is transported through the atmosphere as windblown particulate matter and is deposited onto land and water by wet and dry deposition. Atmospheric loading rates of aluminum to Lake Michigan were estimated at 5 million kg/year (Eisenreich 1980). In this study, most of the aluminum was generally associated with large particles that were deposited near their source. In a recent study, the wet and dry deposition of aluminum was measured biweekly for one year at two sites on Massachusetts Bay, Turro and Nahant. The average total deposition rate was $0.1 \text{ g/m}^2\text{-year}$, of which 29% was in rain (wet deposition) (Golomb et al. 1997).

The mobilization of aluminum by acid rain results in more aluminum being available for plant uptake (Brusewitz 1984). Plant species and cultivars of the same species differ considerably in their ability to take up and translocate aluminum to above-ground parts (Kabata-Pendias and Pendias 1984). Because the tea plant, *Symplocos spicata*, is able to grow in very acidic soils, where aluminum is readily available for uptake by the roots, high concentrations of aluminum may be found in the leaves which serve as a sink for the aluminum (Lewis 1989). Aluminum is often taken up and concentrated in root tissue (Kabata-Pendias and Pendias 1984). In sub-alpine ecosystems, the large root biomass of the Douglas fir, *Abies amabilis*, takes up aluminum and immobilizes it, preventing large accumulation in above-ground tissue (Vogt et al. 1987). It is unclear to what extent aluminum is taken up into root food crops and leafy vegetables. An uptake factor (concentration of aluminum in the plant/concentration of aluminum in soil) of 0.004 for leafy vegetables and 0.00065 for fruits and tubers has been reported (Baes et al. 1984), but the pH and plant species from which these uptake factors were derived are unclear. Based upon these values, however, it is clear that aluminum is not taken up in plants from soil, but is instead biodiluted.

Transfer coefficients of $0.0002 \text{ (kg-day)}^{-1}$ for uptake into milk and $0.0015 \text{ (kg-day)}^{-1}$ for uptake into beef tissue have been reported (Baes et al. 1984). The transfer coefficients represent the fraction of daily aluminum intake in feed that is transferred to a kilogram of milk or beef muscle. Based upon the above values, aluminum is not transferred to beef muscle or milk from feed to any appreciable extent and therefore would not be expected to bioaccumulate in terrestrial food chains.

The potential for accumulation of aluminum has been studied in several aquatic species including fish (Buckler et al. 1995; Cleveland et al. 1991; Hamdy 1993; McDonald et al. 1991; Wilkinson and Campbell 1993), amphibians (Freda and McDonald 1990), crustaceans (Madigosky et al. 1991), snails (Brooks et al. 1992), aquatic insects (Frick and Herrmann 1990; Guerold et al. 1995; Krantzberg and

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Stokes 1990), and aquatic plants (Albers and Camardese 1993; Vuori et al. 1990). Bioconcentration of aluminum in fish is a function of the water quality (e.g., pH and total organic carbon) (Cleveland et al. 1989).

Brook trout have been shown to accumulate slightly more aluminum (measured as whole-body residues) at pH 5.6-5.7 than at pH 6.5-6.6 (Cleveland et al. 1989). Then Cleveland et al. (1991) reported that the estimated steady-state bioconcentration factors (BCF) values for aluminum in brook trout, (which were inversely related to pH), were 215 at pH 5.3, 123 at pH 6.1, and 36 at pH 7.2. The maximum BCFs were 232 at pH 5.3, 153 at pH 6.1, and 46 at pH 7.2. When transferred to water of the same pH without added aluminum brook trout eliminated aluminum from tissues more rapidly at pH 5.3 than at pH 6.1 and 7.2. In tissues of smallmouth bass, aluminum concentrations were higher and more variable in gill tissue than in other tissues (Brumbaugh and Kane 1985). Aluminum concentrations in rainbow trout from an alum-treated lake, an untreated lake, and a hatchery were highest in gill tissue and lowest in muscle (Buerger and Soltero 1983). Aluminum residue analyses in brook trout have shown that whole-body aluminum content decreases as the fish advance from larvae to juveniles (Cleveland et al. 1989). These results imply that the aging larvae begin to decrease their rate of aluminum uptake, to eliminate aluminum at a rate that exceeds uptake, or to maintain approximately the same amount of aluminum while the body mass increases. The decline in whole-body aluminum residues in juvenile brook trout may be related to growth and dilution by edible muscle tissue that accumulated less aluminum than did the other tissues (Cleveland et al. 1989). Wilkinson and Campbell (1993) studied aluminum uptake in Atlantic salmon at a pH of 4.5 under conditions simulating spring snowmelt. These authors reported that gill uptake was slow, approaching a steady state only after 3 days of exposure. The greatest fraction of the gill-associated aluminum was not sorbed to the gill tissue, but to the gill mucus. The authors believe that the mucus appears to retard aluminum transport from solution to the membrane surface, thus delaying the acute biological response of the fish. Most recently, Buckler et al. (1995) reported concentrations of aluminum in whole-body tissue of the Atlantic salmon exposed to high concentrations of aluminum ranging from 3 $\mu\text{g/g}$ (for fish exposed to 33 $\mu\text{g/L}$) to 96 $\mu\text{g/g}$ (for fish exposed to 264 $\mu\text{g/L}$) at pH 5.5. After 60 days of exposure, BCFs ranged from 76 to 190 and were directly related to the aluminum exposure concentration. In acidic waters (pH 4.6-5.3) with low levels of calcium (0.5-1.5 mg Ca/L), labile aluminum between 25 and 75 $\mu\text{g/L}$ is toxic (Rosseland et al. 1990). Because aluminum is toxic to many aquatic species, it is not bioaccumulated to a significant degree ($\text{BCF} < 300$) in most fish and shellfish; therefore, consumption of contaminated fish does not appear to be a significant source of aluminum exposure in humans.

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Aluminum uptake for the leopard frog (*Rana pipiens*) was positively correlated to exposure time and pH; however, no BCF values were reported because the authors felt that the body aluminum accumulation was too variable for useful prediction of the exposure history or physiological status of the frogs (Freda and McDonald 1990).

Bioconcentration of aluminum has also been reported for several aquatic invertebrate species. A BCF value of 0.13-0.5 in the whole-body was reported for the snail, *Helix aspersa*, fed a single 24-hour meal containing aluminum in a barley-flour pellet (Brooks et al. 1992). Madigosky et al. (1991) reported high tissue residues of aluminum in the red swamp crayfish (*Procambarus clarkii*) collected from roadside drainage ditches in Louisiana. Mean aluminum concentrations as $\mu\text{g/g}$ (ppm) dry weight (d/w) in crayfish from roadside ditches ranged from 1.75-6.39 in abdominal muscle, 3.1-22.74 in the hepatopancreas, 309.4-981.50 in the alimentary tract, 10.85-77.45 in the exoskeleton, and 30-140 in the blood. These values were significantly elevated above those of control crayfish where the concentrations ($\mu\text{g Al/g d/w}$) were 1.22 in abdominal muscle, 1.42 in the hepatopancreas, 26.97 in the alimentary tract, 4.28 in the exoskeleton, and 37.9 in the blood.

Bioconcentration of aluminum has also been reported for aquatic insects. Frick and Herrmann (1990) reported aluminum accumulation in mayfly nymphs (*Heptagenia sulphurea*) at low pH (4.5). The nymphs were exposed at 2 concentrations (0.2 and 2 mg inorganic aluminum per liter) and for 2 exposure times (2 and 4 weeks) the longer time period including a molting phase. When nymphs were exposed to the higher concentration of aluminum for 2 instar periods, with a molt in between, the aluminum content (2.34 mg Al/g dry weight) nearly doubled compared with that of a one-instar treatment (1.24 mg Al/g dry weight). The major part of the aluminum was deposited in the exuviae of the nymphs, as the aluminum determination in the nymphs showed a 70% decrease in aluminum content after molting. These authors speculate that internally accumulated aluminum in the nymphs may be transferred to terrestrial predators (e.g., birds). They also hypothesized that externally deposited aluminum may be transferred to terrestrial food chains by aquatic invertebrates that leave the water in their last instar to molt on shore. An important contribution to the idea of biomagnification of aluminum was made by Nyholm (1981). Using semi-quantitative multi-element microanalysis, he related impaired breeding of pied flycatchers (*Ficedula hypoleuca*) in Sweden to the occurrence of aluminum in the bone marrow of the birds. A diet of stoneflies was suspected of forming a link between the lake and the terrestrial predators. Although the matter is far from clear, Nyholm (1981) seems to imply that the insects (stoneflies) were adults and that

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these could contain significant amounts of aluminum even after having left the exuviae behind (Frick and Herrmann 1990).

Vuori et al. (1990) sampled tufts of the aquatic moss *Fontinalis dulecarlica* from the River Lestijoki in Western Finland. The concentrations of aluminum in the water were low (87-196 $\mu\text{g/L}$ [ppb]) due to the high pH values; however, the concentrations in the young terminal shoots of *F. dulecarlica* appeared to be quite high (303-1,852 $\mu\text{g/g}$ [ppm] dry weight). The authors concluded that there was an effective accumulation of aluminum in the moss tissue. Albers and Camardese (1993) compared concentrations of aluminum and other metals in aquatic species of 3 acidified ($\text{pH} \approx 5$) and 3 nonacidified ($\text{pH} \approx 6.5$) constructed wetlands. They found that the metal content of *Sparganium americanum* (bur-reed) was only slightly affected by acidification.

5.3.2 Transformation and Degradation

Because aluminum is an element, its atoms do not degrade in the environment. In addition, aluminum compounds occur in only one oxidation state, $\text{Al}(+3)$. Aluminum can complex with electron-rich species that occur in the environment. The forms of aluminum encountered in a natural system are determined by the strength of the attraction between the positively charged aluminum and the anionic or negatively charged ligands, and the preponderance and types of ligands that are present. These factors will be influenced by pH.

5.3.2.1 Air

Aluminum-containing particulate matter in the atmosphere is mainly derived from soil and industrial processes where crustal material (e.g., minerals) are processed. Aluminum is found as silicates, oxides, and hydroxides in these particles (Eisenreich 1980). Aluminum compounds cannot be oxidized and atmospheric transformations would not be expected to occur during transport. Should aluminum metal particles be released during metal processing, they would be rapidly oxidized.

5.3.2.2 Water

The trivalent aluminum ion is surrounded by six water molecules in solution (Cotton and Wilkinson 1988). The hydrated aluminum ion undergoes "hydrolysis," in which a stepwise replacement of

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coordinated “water of hydration” molecules by hydroxyl ions occurs with the release of a proton from each water molecule into solution (Snoeyink and Jenkins 1980). This results in the formation of hydroxyaluminum species such as $\text{Al}^2(\text{OH}_2)_4$, $\text{Al}(\text{OH})_3$ (insoluble), and $\text{Al}(\text{OH})_4^-$ (Snoeyink and Jenkins 1980). Additional hydrated species such as $\text{Al}_2(\text{OH})_2^{4+}$, $\text{Al}(\text{OH})_5^{-2}$ and hydroxy polymers such as $\text{Al}_{13}(\text{OH})_{32}^{7+}$ have been reported (Bodek et al. 1988; Martell and Motekaitis 1989). The hydrated trivalent aluminum ion is the predominant form at pH levels below 4. Between pH 5 and 6, the predominant hydrolysis products are $\text{Al}(\text{OH})^{2+}$ and $\text{Al}(\text{OH})_2^+$ while the solid $\text{Al}(\text{OH})_3$ is most prevalent between pH 5.2 and 8.8 (Martell and Motekaitis 1989). The soluble species $\text{Al}(\text{OH})_4^-$ is the predominant species above pH 9, and is the only species present above pH 10 (Martell and Motekaitis 1989). Polymeric aluminum hydroxides appear between pH 4.7 and 10.5, and increase in size until they are transformed into colloidal particles of amorphous $\text{Al}(\text{OH})_3$ which crystalize to gibbsite in acid waters (Brusewitz 1984). Polymerization is affected by the presence of dissolved silica; when enough silica is present, aluminum is precipitated as poorly crystallized clay mineral species (Bodek et al. 1988).

Hydroxyaluminum compounds are considered “amphoteric” (e.g., they can act as both acids and bases in solution) (Cotton and Wilkinson 1988). Because of this property, aluminum hydroxides can act as buffers and resist pH changes within the narrow pH range of 4-5 (Brusewitz 1984).

Monomeric aluminum compounds, typified by aluminum fluoride, chloride, and sulfate, are considered reactive or labile compounds, whereas polymeric aluminum species react much more slowly in the environment (Hemenway and Fitzgerald 1984). Aluminum has a stronger attraction for fluoride in an acidic environment compared to other inorganic ligands (Brusewitz 1984), and fluoride complexes of aluminum have been shown to be more toxic to fish than aluminum-organic complexes are (Plankey and Patterson 1987). Fulvic acid is also an important ligand for aluminum under acidic conditions, and it has been observed that as the temperature is lowered, the rate of complexation of aluminum with fluoride is considerably slowed, while the rate of complexation between aluminum and fulvic acid is only slightly decreased in rate (Plankey and Patterson 1987). This suggests that during snow-melt conditions, when aluminum and hydrogen ion concentrations increase, complexation with fulvic acid could preferentially occur over complexation with fluoride.

5.3.2.3 Sediment and Soil

Aluminum is present in many primary minerals. The weathering of these primary minerals over time results in the deposition of sedimentary clay minerals, such as the aluminosilicates kaolin and montmorillonite. The weathering of soil results in the more rapid release of silicon, and aluminum precipitates as hydrated aluminum oxides such as gibbsite and boehmite, which are constituents of bauxites and laterites (Bodek et al. 1988). Aluminum is found in the soil complexed with other electron rich species such as fluoride, sulfate, and phosphate.

5.4 LEVELS MONITORED OR ESTIMATED IN THE ENVIRONMENT

Reliable evaluation of the potential for human exposure to aluminum depends in part on the reliability of supporting analytical data from environmental samples and biological specimens. In reviewing data on aluminum levels monitored or estimated in the environment, it should also be noted that the amount of chemical identified analytically is not necessarily equivalent to the amount that is bioavailable. The analytical methods available for monitoring aluminum in various environmental media are detailed in Chapter 6.

5.4.1 Air

Aluminum is found in the atmosphere mainly as aluminosilicates associated with crustal particulate matter. There are varying levels of aluminum in the atmosphere, depending on the location of the sampling site, meteorologic conditions, and the level of industrial activity or traffic in the area. Aluminum levels are expected to be low in areas influenced by the ocean and high in areas with wind-blown soil. Background levels of aluminum in the atmosphere generally range from 0.005 to 0.18 ng/m³ (Hoffman et al. 1969; Poetzl 1970; Sorenson et al. 1974). In rural areas of Hawaii, aluminum concentrations have been measured at a range of 0.005-0.032 ng/m³ (Hoffman et al. 1969), whereas a concentration range of 0.27-0.39 ng/m³ has been reported in Manitoba National Park in Canada (Rahn 1971). Atmospheric aluminum concentrations in U.S. cities and industrial areas are considerably higher, ranging from about 0.4 to 10 ng/m³ (Cooper et al. 1979; Dzubay 1980; Kowalczyk et al. 1982; Lewis and Macias 1980; Moyers et al. 1977; Ondov et al. 1982; Pillay and Thomas 1971; Sorenson et al. 1974; Stevens et al. 1978). The range of the concentration of aluminum in fine (<1-2.5 μm) and coarse (2.5-10 μm) particles from two industrial areas, Southeast Chicago and East St. Louis were 22-539 ng/m³

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(125 ng/m³ mean) and 24-1370 ng/m³ (153 ng/m³ mean), respectively for fine particles and 8.2-1760 ng/m³ (390 ng/m³ mean) and 17-2120 ng/m³ (442 ng/m³ mean), respectively for coarse particles. At a rural site (Bondville, IL), the aluminum concentrations in fine and coarse particles were 32-293 ng/m³ (95 ng/m³ mean) and 32-3120 ng/m³ (338 ng/m³ mean), respectively which was not much different than the aluminum concentration from the industrial sites (Sweet et al. 1993).

Aluminum levels can also vary with seasonal meteorological conditions. For example, in Mackinac Island, Michigan, summer levels averaged about 0.25 ng/m³, while winter levels were only about 0.18 ng/m³ (Rahn 1971).

Aluminum has been identified in air samples collected at 9 of the 427 NPL hazardous waste sites where it has been detected in some environmental media (HazDat 1996).

5.4.2 Water

The concentrations of dissolved aluminum in water vary with pH and the humic-derived acid content of the water (Brusewitz 1984). Aluminum is only sparingly soluble in water between pH 6 and pH 8. Because the pH of about 95% of naturally-occurring water is between 6 and 9 and since high aluminum concentrations occur in surface water bodies only when the pH is < 5, the aluminum concentration in most natural waters is extremely low (Filipek et al. 1987; Snoeyink and Jenkins 1980; Sorenson et al. 1974). In general, aluminum concentrations in surface waters at pH levels above 5.5 will be < 0.1 mg/L (ppm) (Brusewitz 1984; Miller et al. 1984; Sorenson et al. 1974; Taylor and Symons 1984). However, even at neutral pH levels, higher aluminum levels have been found in lakes with a high humic acid content (Brusewitz 1984). At lower pH levels, the aluminum content significantly increases because of increased solubility of aluminum oxide and salts in acidic solutions. For example, aluminum has been found at concentrations of up to 90 mg/L (ppm) in tributaries that drain mines containing massive sulfide deposits (Filipek et al. 1987). In heavily contaminated surface waters in a mining region rich in sulfides, the water was highly acidic (pH < 3.5) and the levels of soluble aluminum were greater than 2 mmol/L (50 mg/L) (Alvarez et al. 1993). Similarly, surface water samples contaminated with acidic mine drainage collected at seven different locations in the vicinity of abandoned coal mines in west-central Indiana had aluminum levels of 6.0 to 269 mg/L (Allen et al. 1996). The pH ranged from 2.1 to 3.4 at these sites.

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Aluminum was detected at dissolved aluminum concentrations ranging from 0.001 to 2.760 mg/L (ppm) with a mean concentration of 0.074 mg/L (ppm) in 456 of 1,577 raw surface water samples collected during a 5-year survey at various locations across the United States (Kopp and Kroner 1970). Dissolved aluminum concentrations were detected in about 48% of the 380 finished drinking waters sampled and ranged from 0.003 to 1.6 mg/L (ppm) with a mean of 0.179 mg/L (ppm) (Kopp and Kroner 1970). In another survey of 186 community water systems, median aluminum concentrations for all finished drinking water samples ranged from 0.026 to 0.161 mg/L (ppm), while the maximum and minimum levels were 2.67 mg/L (ppm) and 0.051 mg/L (ppm) respectively (Miller et al. 1984). These authors further reported that the median aluminum concentration in finished water that received no coagulation treatment was 0.043 mg/L (ppm) (range, 0.016-1.167 mg/L) compared to the median of 0.112 mg/L (ppm) (range, 0.014-2.670 mg/L) in finished water receiving alum (aluminum sulfate) coagulation treatment. In the supplies in which no coagulant was used during treatment, 29% of supplies using surface water as their source had aluminum levels exceeding 0.05 mg/L, whereas only 4% of supplies using groundwater sources exceeded this level. When aluminum coagulants were used, 69% of all supplies had residual aluminum concentrations greater than 0.05 mg/L. In another study, the aluminum content in treated water at facilities using alum coagulation treatment of raw waters ranges from about 0.01 to 1.3 mg/L (ppm) with a mean of about 0.157 mg/L (ppm) (Letterman and Driscoll 1988).

Most recently, Schenck et al. (1989) measured aluminum concentrations in drinking water collected primarily in the western and central parts of the United States from outlets from which water was consumed rather than from the original water treatment plant (Table 5-3). Although aluminum levels in household tap water may range from 0 to 1.029 mg/L (ppm), aluminum levels in most drinking water in the United States were <0.1 ppm (Schenck et al. 1989). While several water sources in the west coast states (California, Oregon, and Washington) were found to contain undetectable levels of aluminum (<0.001 ppm), several cities in other geographic areas of the U.S. had high aluminum concentrations (>0.4 ppm). These included Peoria, Illinois (0.467 ppm); Coos Bay, Oregon (0.483 ppm); Watertown, South Dakota (0.502 ppm); Waco, Texas (0.520 ppm); Yellowstone National Park, Wyoming (0.608 ppm); Philadelphia, Pennsylvania (0.688 ppm); and Charleston, South Carolina (1.029 ppm).

Aluminum has been measured in atmospheric precipitation (i.e., rain and snow) in the United States at concentrations up to 1.2 mg/L (ppm) (Dantzman and Breland 1970; Feth et al. 1964; Fisher et al. 1968; Norton 1971). Most recently, aluminum has been measured in rainwater samples collected on-board

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Table 5-3. Aluminum Concentrations Detected in Drinking Water in Various Regions of the United States

U.S. States	Aluminum Concentration (ppb) ^a
California	0-274
Colorado	42-166
Hawaii	12-124
Idaho	28-63
Illinois	3-467
Indiana	1-137
Kansas	12-245
Kentucky	9-400
Louisiana	12-210
Michigan	6-123
Minnesota	24-93
Missouri	2-368
Montana	11-98
New York ^b	254-299
Nevada	5-126
Ohio	2-245
Oregon	0-483
Pennsylvania ^c	688
South Carolina	2-1,029
South Dakota	2-502
Tennessee ^d	45
Texas	1-520
Utah	19-51
Washington	0-118
Wisconsin	12-118
Wyoming	16-608

Source: Schenk et al. 1989

^aRange in values reported for each state

^bWater sampled in New York City only

^cWater sampled in Philadelphia only (one sample)

^dWater sampled in Memphis only (one sample)

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ship during the Global Change Expedition in the North Atlantic Ocean (Lim and Jickells 1990). These authors reported that comparisons between acid-leachable and total (dissolved plus particulate) trace aluminum concentrations suggest that the acid-leachable fraction of aluminum can significantly underestimate total concentrations of aluminum in rainwater. Acid-leached mean concentrations of aluminum in rainwater collected during 3 rainfall events in the North Atlantic were 33.7, 12.2, and 1.99 $\mu\text{g/L}$ (ppb). Overall, the acid-leached concentrations of aluminum in rainwater for seven rainfall events ranged from 1.14 to 35.2 $\mu\text{g/L}$ (ppb). These values were compared with acid-leachable aluminum concentrations in precipitation from remote areas which ranged from 2.1 to 15.44 $\mu\text{g/L}$ (ppb) (Lim and Jickells 1990). Total (dissolved plus particulate) aluminum concentrations in North Atlantic precipitation samples collected in 1988 ranged from 6.1 to 824 $\mu\text{g/L}$ (ppb). A comparison with atmospheric aluminum concentrations presented previously indicates that one liter of precipitation cleanses the aluminum from an equivalent of 0.5 to 7 million cubic meters of air.

Aluminum levels in marine waters tend to be much lower (i.e., <0.001 mg/L [<1 ppb]) than those found in fresh water lakes and streams (Brusewitz 1984), probably because of increased alkalinity in marine waters compared to fresh waters.

Aluminum levels in groundwater wells at neutral pH generally fall below 0.1 mg/L (100 ppb) (Brusewitz 1984). In areas receiving acid precipitation, aluminum levels in groundwater may be more than 10 times the levels found in areas with neutral pH levels in the water (Brusewitz 1984), possibly due to precipitation of aluminum compounds in the more alkaline medium or the reaction of aluminum with available silicates. In another study, Miller et al. (1984) reported that the median concentration of aluminum in finished water obtained from groundwater was 0.031 mg/L (ppm) (range, 0.014-0.290 mg/L) as compared to the median concentration in surface water of 0.043 mg/L (ppm) (range, 0.016-1.167 mg/L). These authors also reported that, while 55% of the raw surface waters sampled contained aluminum concentrations >0.05 mg/L , only 4% of the raw groundwater samples contained aluminum concentrations >0.05 mg/L (ppm).

Aluminum has been identified in surface water, leachate, and groundwater samples collected at 227,54, and 336 of the 427 NPL hazardous waste sites, respectively, where it has been detected in some environmental media (HazDat 1996).

5.4.3 Sediment and Soil

Aluminum is the third most abundant element and the most common metal in the earth's crust, comprising about 8% of the lithosphere (Lide 1997). Its concentration in soils varies widely, ranging from about 0.07 percent by weight or 700 mg/kg (ppm) to over 10 percent by weight or 100,000 mg/kg (ppm) (Shacklette and Boerngen 1984; Sorenson et al. 1974). Data gleaned from texts and literature reviewed by soil scientists suggest a typical aluminum concentration in soil as 71,000 mg/kg (Frink 1996). Varying concentrations are found in different soil samples taken from the same area and in areas with different vegetation types (Brusewitz 1984; Sorenson et al. 1974). For example, in different soils of Missouri, aluminum concentrations ranged from 4,800 to 58,000 mg/kg (ppm) (USGS 1972). In Hawaii, aluminum contents were much higher with concentrations ranging from 79,000 to 317,000 mg/kg (ppm) (Moomaw et al. 1959). Soils in Florida and parts of Georgia, Texas, Oklahoma, and Michigan contain less than 20,000 mg/kg of soil, whereas soils from portions of the Pacific Northwest, New England, Colorado, and Nevada have concentrations greater than 80,000 mg/kg (Sparling and Lowe 1996). The aluminum content in cultivated and uncultivated soil samples collected during a number of field studies ranged from 7,000 mg/kg to over 100,000 mg/kg (ppm) (mean concentration of 33,000 mg/kg) for subsurface soils in the eastern United States, from 5,000 mg/kg to over 100,000 mg/kg (ppm) (mean concentration of 54,000 mg/kg) for subsurface soils in the western United States, and from 13,000 to 76,000 mg/kg (ppm) for surface horizon soils collected in Colorado (mean concentration of 57,000 mg/kg) (Connor and Shacklette 1975). The aluminum content of soils is strongly correlated with its clay content (Ma et al. 1997).

Aluminum levels in soil also vary with different vegetation types. For example, aluminum levels in the soils of coniferous forests are often higher than in soils of beech forests since coniferous forests tend to have more acid soils (Brusewitz 1984). Alternate views of the data are that the acidic soil produced by conifers can preferentially mobilize aluminum from deeper layers toward surface soil, or that conifers over beech preferentially grow in soils rich in aluminum and it is their metabolic processes which produce more acidic soil. An analysis of aluminum in soils by depth could improve the understanding of this process.

Aluminum has been identified in soil and sediment samples collected at 203 and 151 of the 427 NPL hazardous waste sites, respectively, where it has been detected in some environmental media (HazDat 1996).

5.4.4 Other Environmental Media

Aluminum occurs naturally in many edible plants and is added to many processed foods. The concentrations in foods and beverages vary widely, depending upon the food product, the type of processing used, and the geographical areas in which food crops are grown (Brusewitz 1984; Sorenson et al. 1974). In general, the foods highest in aluminum are those that contain aluminum additives (e.g., processed cheese, grain products, and grain-based desserts) (Greger 1992; Pennington 1987). Because of the variability of reported levels of aluminum in foods, the many new manufactured food products on the market, and the increasing use of aluminum as a packaging material, a wide range of beverages and foods have been analyzed. The aluminum concentrations in a number of beverages, foods, and food products are listed in Table 5-4. Most unprocessed foods, (with the exception of some herbs and tea leaves) typically contain less than 5 mg/kg (ppm) aluminum (Greger 1992). Furthermore, only small quantities of herbs are consumed by most individuals, and most of the aluminum in tea leaves is in an insoluble form. The measured levels of aluminum in unprocessed foods range from about 0.1 mg/kg (ppm) in eggs, apples, raw cabbage, corn, and cucumbers to 7.16 mg/kg (ppm) in lettuce (Schenck et al. 1989). Unregulated and unanalyzed natural dietary supplements represent an uncertain introduction of aluminum into the diet.

It should be noted, however, that the aluminum content of some plants known to be aluminum accumulators can vary greatly, depending on the plant variety and soil conditions, including pH in which it is grown (Greger 1992). Preliminary data indicate that plants grown on soil amended with a low weight percentage of ash from power plants take up aluminum to a higher extent than from unamended soil (Bathe et al. 1991). This suggests that aluminum ash is more available to plants than that in ordinary soil and that the aluminum content in plants will be affected by the nature of aluminum-containing amendments, both intentional and unintentional, to soil. The broad variation in the occurrence of aluminum in food plants is exemplified in the tea plant. The aluminum content in tea (1% extract) usually ranges from 0.378 to 2.445 mg/L (ppm). Because the tea plant is able to grow in very acidic soils, where aluminum is readily available for uptake by the roots, the tea leaves serve as a sink, accumulating up to 10,000 mg/kg (ppm) (Lewis 1989). However, herbal tea contains lower levels of aluminum than ordinary tea (0.140-1.065 mg/L [ppm]) (Schenck et al. 1989). The aluminum content in ash samples from other cultivated plants (e.g., lima beans, cabbage, soybeans, and tomatoes) collected during a number of field studies in Georgia, Missouri, and Wisconsin ranged from 50 to 30,000 mg/kg (ppm) (mean concentration range: 200-1,700 mg/kg [ppm]) with the highest levels

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Table 5-4. Estimated Aluminum Concentrations of Selected Foods

Foods	Aluminum concentration ($\mu\text{g/g}$)	Reference
Beverages (mg/L)		
Fruit juices (e.g., orange, reconstituted lemon, peach)	0.043–4.130	Derived from Schenk et al. 1989
Soft drinks (e.g., ginger ale, diet cola)	0.103–2.084	Derived from Schenk et al. 1989
Cola, carbonated	0.1	Pennington 1987
Alcoholic beverages (e.g., beer, wine, wine coolers, champagne)	0.067–3.20	Derived from Schenk et al. 1989
Beer, canned	0.07	Pennington 1987
Spirits (e.g., brandy, vodka, whiskey)	0.148–0.635	Derived from Schenk et al. 1989
Tea, steeped from tea bags	0.424–2.931	Derived from Schenk et al. 1989
Herbal teas (1% extract)	0.14–1.065	Derived from Schenk et al. 1989
Tea, steeped	4.3	Greger et al. 1985b
Instant coffee (1% solution)	0.02–0.581	Derived from Schenk et al. 1989
Whole coffee (3% extract)	0.235–1.163	Derived from Schenk et al. 1989
Animals Products		
Beef, cooked ^a	0.2 ^b	Greger et al. 1985b
Cheese (e.g., Swiss, cheddar, bleu)	3.83–14.10	Derived from Schenk et al. 1989
Cheese, cheddar	0.2	Pennington 1987
Cheese, cottage, creamed	0.1	Pennington 1987
Cheese, processed	297 ^b	Greger et al. 1985b
Chicken, with skin, cooked ^a	0.7	Greger et al. 1985b
Egg	0.107	Derived from Schenk et al. 1989
Eggs, scrambled	2.865	Derived from Schenk et al. 1989
Eggs, cooked ^a	0.1	Greger et al. 1985b
Fish (cod), cooked ^a	0.4	Greger et al. 1985b
Fish, salmon	5.44	Derived from Schenk et al. 1989
Fish, herring	0.127	Derived from Schenk et al. 1989
Ham, cooked ^a	1.2	Greger et al. 1985b

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Table 5-4. Estimated Aluminum Concentrations of Selected Foods (continued)

Foods	Aluminum concentration ($\mu\text{g/g}$)	Reference
Milk, whole	0.06	Pennington 1987
Milk (skim, whole, and powdered)	0.102–1.409	Derived from Schenk et al. 1989
Salami	1.1	Pennington 1987
Yoghurt, plain low-fat	1.1	Pennington 1987
Fruits		
Apple	0.1	Pennington 1987
Banana, fresh	0.05	Pennington 1987
Grapes	0.5 ^b	Sorenson et al. 1974
Orange juice, frozen reconstituted	0.06	Pennington 1987
Peaches	0.4 ^b	Sorenson et al. 1974
Raisins, dried	3.1	Pennington 1987
Strawberries, fresh	2.2	Pennington 1987
Grains		
Biscuits, baking powder, refrigerated	16.3	Pennington 1987
Bread, white	3	Sorenson et al. 1974
Bread, white	0.351	Derived from Schenk et al. 1989
Bread, pumpernickel	13.2	Derived from Schenk et al. 1989
Bread, whole wheat	5.4	Sorenson et al. 1974
Cereal (e.g., Post Raisin Bran®, Malt-o-Meal Wheat Cereal®)	0.040–29.33	Derived from Schenk et al. 1989
Corn chips	1.2	Pennington 1987
Cornbread, homemade	400	Pennington 1987
Muffin, blueberry	128	Pennington 1987
Oatmeal, cooked	0.7	Pennington 1987
Oats	2.21–4.18	Derived from Schenk et al. 1989

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Table 5-4. Estimated Aluminum Concentrations of Selected Foods (continued)

Foods	Aluminum concentration ($\mu\text{g/g}$)	Reference
Rice, cooked ^a	1.7	Greger et al. 1985b
Rice, yellow, Rice-a-Roni®	1.97	Derived from Schenk et al. 1989
Spaghetti, cooked ^a	0.4	Greger et al. 1985b
Vegetables and Legumes		
Asparagus	4.4 ^b	Schlettwein-Gsell and Mommsen-Straub 1973
Beans, green, cooked ^a	3.4	Greger et al. 1985b
Beans, navy, boiled	2.1	Pennington 1987
Cabbage, raw	0.1	Greger et al. 1985b
Cauliflower, cooked	0.2	Greger et al. 1985b
Corn	0.1	Pennington 1987
Cucumber, fresh, pared	0.1	Pennington 1987
Lettuce	0.6	Schlettwein-Gsell and Mommsen-Straub 1973
Lettuce	7.16	Derived from Schenk et al. 1989
Peanut butter	5.8	Pennington 1987
Peanut butter, natural	6.29	Derived from Schenk et al. 1989
Peas, frozen, Pict Sweet®	1.64	Derived from Schenk et al. 1989
Peas, green, cooked	1.9	Greger et al. 1985b
Potatoes, unpeeled, boiled ^a	0.1	Greger et al. 1985b
Potatoes, unpeeled, baked	2.4	Greger et al. 1985b
Potato, red	3.63	Derived from Schenk et al. 1989
Potato, sweet	1.01	Derived from Schenk et al. 1989
Spinach, cooked ^a	25.2 ^b	Schlettwein-Gsell and Mommsen-Straub 1973
Tomatoes, cooked ^a	0.1	Greger et al. 1985b
Herbs and Spices		
Basil	3,082 ^b	Sorenson et al. 1974

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Table 5-4. Estimated Aluminum Concentrations of Selected Foods (continued)

Foods	Aluminum concentration ($\mu\text{g/g}$)	Reference
Celery seed	465 ^b	Sorenson et al. 1974
Cinnamon	82 ^b	Sorenson et al. 1974
Oregano	600 ^b	Sorenson et al. 1974
Pepper, black	143 ^b	Sorenson et al. 1974
Thyme	750 ^b	Sorenson et al. 1974
Other Food Products		
Baking powder	2,300 ^b	Sorenson et al. 1974
Candy, milk chocolate	6.8	Pennington 1987
Chocolate cookie, Oreo	12.7	Derived from Schenk et al. 1989
Cocoa	45	Schlettwein-Gsell and Mommsen-Straub 1973
Cream substitute, powdered	139	Pennington 1987
Nondairy creamer	25.7–94.3	Derived from Schenk et al. 1989
Pickles with aluminum additives	39.2 ^b	Greger et al. 1985b
Pickles	0.126–9.97	Derived from Schenk et al. 1989
Salad dressing, Kraft Miracle Whip®	3.7	Derived from Schenk et al. 1989
Salt with aluminum additives	164 ^b	Greger et al. 1985b
Salt	31.3–36.6	Derived from Schenk et al. 1989
Soup	0.032–3.6	Derived from Schenk et al. 1989

^aFood reported to *not* be stored or cooked in aluminum pans, trays, or foil.

^bValue is an average of several values reported in the reference.

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occurring in cabbage and lima beans from Georgia and the lowest levels occurring in Missouri soybeans and Georgia tomatoes (Connor and Shacklette 1975). The aluminum content in ash samples from corn in Georgia, Missouri, and Wisconsin ranged from 50 to 3,000 mg/kg (ppm) with mean concentrations ranging from 200 to 1,000 mg/kg (ppm) (Connor and Shacklette 1975).

In fiscal years 1985/1986, the FDA conducted a survey of elements in fresh clams and oysters collected from U.S. coastal areas in use for shellfish production (Capar and Yess 1996). The average concentration (wet weight basis) of aluminum found in the four shellfish categories surveyed were: clams (hardshell), 23 ± 23 mg/kg (n=74); clams (softshell), 115 ± 110 mg/kg (n=59); Eastern oyster, 33 ± 26 mg/kg (n=104); Pacific oyster, 30 ± 28 mg/kg (n=46). Cod and bluefin tuna from the Northwest Atlantic Ocean contained an average of 1 and 0.4 mg/kg of aluminum respectively, in muscle tissue (Hellou et al. 1992a, 1992b).

The high aluminum concentrations seen in some processed foods (e.g., processed cheeses, baked goods, and nondairy cream substitutes) are likely to have been introduced into the foods as additives, such as the anti-caking agent, sodium aluminosilicate, which is present in salt, non-dairy creamers, and many other powdered materials (Table 5-4) (Schenck et al. 1989). The most commonly used food additives containing aluminum are: acidic sodium aluminum phosphate (leavening agent in baked goods); basic sodium aluminum phosphate (emulsifying agent in processed cheese); aluminum sulphates (acidifying agents); bentonite (materials-handling aid); aluminum color additives (lakes) from various food dyes, and aluminum silicates (anti-caking agents) (Greger 1992).

Aluminum has also been found in infant milk formulas although it is not clear whether it is contained in one of the ingredients or has been introduced during processing (Koo et al. 1988; Simmer et al. 1990; Weintraub et al. 1986). Aluminum levels were measured in 175 samples of whole milk, milk formulas, and other nutrient products commonly used for infants as part of a study of the possible relationship between ingested aluminum and bone disorders. Aluminum content was lowest in human milk, various cow milk preparations, bottled sterile water and glucose water, and most oral multivitamin preparations. Aluminum levels were highest in modified infant formulas, including soy formula, premature infant formula, and products for specific metabolic disorders (Koo and Kaplan 1988; Simmer et al. 1990; Weintraub et al. 1986). The mean concentration of aluminum in U.S. infant milk formulas has been reported to range from 0.14 to 3.74 mg/L (ppm) for liquid formulas and from 6.25 to 11.8 mg/kg (ppm) for powdered formulas (Weintraub et al. 1986). This corresponds to an aluminum content of

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0.125-1.89 ppm of feed for liquid formulas and 0.935-1.78 ppm of feed for powdered formulas.

Compared with a liter of breast milk (0.012-0.147 ppm) (Simmer et al. 1990), the aluminum content per liter of reconstituted formula (i.e., diluted according to the manufacturer's recommendations) was up to 63 times greater (Weintraub et al. 1986). These authors also reported that infant formulae from other countries contained up to 165 times the aluminum content of breast milk or cows milk. A more recent study in the United Kingdom found aluminum levels in cows' milk, soy milk, and human breast milk in the range of 4-33 µg/L (14 µg/L mean), 5 to 285 µg/L (160 µg/L mean), and 3 to 79 µg/L (27 µg/L mean), respectively (Baxter et al. 1991). Mean aluminum concentrations in the soy and cows' milk-based samples were, on average, 37% and 45% lower, respectively, than the same brands purchased between 1985 and 1987. The authors also surveyed 1990 retail samples of infant formula. The estimated concentration of aluminum in the prepared feed ranged from 530 to 640 µg/L in soy-based formula and 27 to 120 µg/L in cows' milk-based formula. Aluminum levels in breast milk, humanized infant formulae, and in special purpose infant formulae are summarized in Table 5-5.

Cooking foods in aluminum pots and pans or storing foods in aluminum foil or cans may increase the aluminum content in some foods since aluminum may dissolve when in contact with a salty, acidic, or alkaline food (Abercrombie and Fowler 1997; Greger et al. 1985b; King et al. 1981; Muller et al. 1993b; Nagy and Nikdel 1986). Table 5-6 compares the concentrations of aluminum in a variety of foods prepared in aluminum cookware as compared to stainless steel cookware. Aluminum concentrations in precooked foods (e.g., applesauce, green beans, beef, eggs, ham pudding, rice, and tomato sauce) ranged from < 0.1 to 21.6 mg/kg (ppm), while concentrations in the foods after cooking in conditioned aluminum pans and stainless steel pans ranged from 0.24 to 125 mg/kg (ppm) and from < 0.1 to 3.4 mg/kg, respectively (Greger et al. 1985b). In the Greger et al. (1985b) study, some foods seemed to readily accumulate aluminum when cooked in aluminum rather than stainless steel. Ranked in order of increasing migration, foods that accumulated aluminum when cooked in aluminum rather than stainless steel pans were: grits, cauliflower, beef, eggs, cabbage, applesauce, and tomato sauce. Acidic foods, such as tomatoes, tomato sauce, and applesauce, especially when cooked for more than 15 minutes, tended to accumulate more aluminum than other foods (Greger et al. 1985b). Greger et al. (1985b) also reported that foods cooked in new aluminum cookware had higher aluminum concentrations than foods cooked in old aluminum cookware or aluminum cookware that had been treated to simulate use. In addition, the aluminum levels in the foods prepared in any aluminum cookware (old, new, or treated to simulate use) had higher aluminum levels than the same foods cooked in stainless steel cookware. Previous analyses suggested that the use of an aluminum pot to prepare tomato sauce could add up to

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Table 5-5. Comparison of Aluminum Levels ($\mu\text{g/L}$) in Breast Milk, Humanized Infant Formulae, and in Special Purpose Infant Formulae

	Mean (\bar{x})	Range ^a
Breast milk	49	12–147
Humanized infant formulae		
Nan powder	1,463	1,201–1,960
Nan ready to feed	1,218	
Lactogen powder	335	77–827
Lactogen liquid	470	366–627
Lactogen ready to feed	248	
S26 powder	192	120–370
S26 powder/sachet	140	92–165
S26 ready to feed	311	
SMA powder	113	103–130
S26 Progress powder/sachet	165	
Enfalac powder	201	188–210
Enfalac ready to feed	350	
Enfalac (reduced iron) powder	246	238, 254 ^b
Enfamil powder	112	95, 132 ^b
Similac powder	72	70, 74 ^b
Karitane powder	448	447, 448 ^b
Karitane follow-on powder	363	
Special purpose infant formulae		
Preterm formulae		
Alprem powder	184	
S26 low birth weight ready to feed	275	
Premature Enfalac powder	337	240, 434 ^b
Premature Enfamil ready to feed	1,106	919–1,312
Enfamil breast milk fortifier		
0.96 g/25 mL water	134	
0.96 g/25 mL breast milk	171	122–207
Soy formulae		
Isomil liquid	1,238	
Isomil powder	1,192	
Infa Soy powder	1,670	
Prosobee powder	1,711	1,613–1,861
Special formulae		
Pregestimil powder	939	846, 1,031 ^b
Nutamigen powder	835	
Alfare powder	456	451, 460 ^b
Portagen powder	493	
Delact powder	62	
Digestelac powder	82	
Karitane goat's milk	360	

Source: Simmer et al. 1990

^aRange provided where three or more batches of milk or infant formulae were analyzed^bOnly two batches of formulae were tested

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Table 5-6. Concentrations of Aluminum (ppm wet weight) in Foods Before and After Cooking in Aluminum^a and Stainless Steel Cookware

Food	Uncooked	Cooked in aluminum cookware	Cooked in stainless steel cookware
Apple sauce	0.13	7.1 ^b	0.12
Beans, green	3.8	3.8	3.4
Beef, rump roast	0.19	0.85 ^b	0.21
Cabbage	0.13	3.6 ^b	0.20
Cauliflower	0.19	0.72	0.19
Chicken	0.47	1.00	0.66
Cod	0.35	0.47	0.40
Eggs	0.10	1.6 ^b	0.13
Grits	0.62	0.60	0.17
Ham	0.85	1.2	1.2
Peas	1.9	1.9	1.9
Pudding	21.3 ^c	4.2	4.0
Rice	1.5	1.7	1.7
Spaghetti	1.7	0.78	0.45
Tomato sauce	0.10	57.1	0.16

Source: Greger et al. 1985b.

^aAluminum pans conditioned through standardized cooking procedures

^bProducts cooked in aluminum pans contained significantly ($p < 0.05$) more aluminum than unprocessed product or product cooked in stainless steel pans.

^cDry product had significantly ($p < 0.05$) higher aluminum concentration than either cooked product.

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4 mg aluminum to each serving of the sauce (Lione 1983). In a small sampling of canned drinks stored at 15-20 °C, the aluminum content ranged from less than 0.1 to 74 ppm depending on the product and storage time (Abercrombie and Fowler 1997). The study concluded that there appeared to be little basis for concern about the ingestion of aluminum when the internal protective coating of cans remains intact, the cans are stored properly, and the contents are consumed in a reasonable period of time.

It has been estimated that brewing coffee in a new aluminum pot can add from 0.88 mg (immediately after brewing) to 1.18 mg aluminum (after a further 12-hour storage in the pot and subsequent reheating) to each cup (Lione et al. 1984). Percolators that have been used repeatedly are less susceptible to mobilization of aluminum by coffee, and brewing in these increases the aluminum content of each cup of coffee by only 0.4 mg immediately after brewing and by 0.58 mg after storage for 12 hours in the pot and reheating. The aluminum content of ground coffee beans has been measured at 51.8 mg/kg (ppm) (Lione et al. 1984).

Muller et al. (1993b) reported migration of aluminum from aluminum cans (unlacquered) into Coca-Cola® (pH 2.5) and diet Coca-Cola® (pH 3.0), and that the concentration of aluminum increased as the storage period increased. Concentrations of aluminum ranged from 46 to 170 µg/L (ppb) in Coca-Cola® (storage for 40-101 days) and from 14 to 250 µg/L (ppb) in diet Coca-Cola® (storage for 44-173 days), respectively. These authors also assessed the migration of aluminum from aluminum cans into 0.08% nitric acid solutions. As was shown for Coca-Cola® 's unlacquered cans, the total amount of aluminum that migrated into the nitric acid solutions increased with increasing storage period.

Aluminum compounds are also used extensively in the manufacture of cosmetics (e.g., aluminum hexahydrate in deodorants) and in medical treatments (e.g., aluminum hydroxide in antacids to control gastric hyperacidity or aluminum oxide in dental ceramic implants) (Brusewitz 1984; NRC 1982). In addition, antacids and buffered aspirin contain 4-562 mg/kg (ppm) of aluminum (Schenck et al. 1989; Shore and Wyatt 1983). Lione (1985a) reported aluminum content/dose (single tablet or 5 mL liquid) for antacids, internal analgesics (buffered aspirins), antidiarrheals, and anti-ulcerative drugs (Table 5-7). The aluminum content per dose (single tablet or 5 mL liquid) ranged from 35 to 208 mg for antacids, 9-52 mg for buffered aspirins, 36-1,450 mg for antidiarrheal drugs, and 207 mg for an anti-ulcerative drug. Potential daily aluminum dosage ranged from 126 to 5,000 mg for these medications.

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Table 5-7. Aluminum-containing Non-prescription Drugs and Sucralfate*

Drug class	Aluminum salts used	Aluminum content/ dose** (mg)	Possible daily dose Al (mg)
1. Antacids	a. aluminum hydroxide	35–208	840–5,000
	b. dihydroxyaluminum acetate	45–72	
	c. aluminum carbonate	n.a.	
	d. aluminum oxide	41	
	e. bismuth aluminate	55	
	f. magaldrate	51–61	
	g. dihydroxyaluminum aminoacetate	100	
	h. dihydroxyaluminum sodium carbonate	63	
2. Internal analgesics (buffered aspirins)	a. aluminum hydroxide	9–52	126–728
	b. aluminum glycinate	35,717	
3. Antidiarrheals	a. kaolin	120–1,450	
	b. aluminum magnesium silicate	36	
	c. attapulgite	500–600	
4. Anti-ulcerative	a. aluminum sucrose sulfate	207	828

* Data modified from Lione 1985a

** Single tablet or 5 mL liquid

Brand name (manufacturer) for the aluminum salts used to each drug class:

1a. Albicon (Pfeiffer), AlternaGel (Stuart), Aludrox (Wyeth), Aluminum Hydroxide Gel (Philips Roxane), Alurex (Rexall), Amphojel (Wyeth), A.M.T. (Wyeth) Antacid Powder (DeWitt), Banacid (Buffington), Basaljel Extra Strength (Wyeth), Camalox (Rorer), Creamalin (Winthrop), Delcid (Merrell-Dow), Dialume (Armour), Di-Gel (Plough), Estomul-M (Riker), Flacid (Amfre-Grant), Gaviscon (Marion), Gaviscon-2 (Marion), Gelumina (Amer. Pharm.), Gelusil (Warner-Chilcott), Gelusil II (Warner-Chilcott), Gelusil M (Warner-Chilcott), Glycogel (Central Pharm.), Kessadrox (McKesson), Kolantyl (Merrill-Dow), Kudrox (Kremers-Urban), Liquid Antacid (McKesson), Maalox (Rorer), Maalox No. 1 (Rorer), Maalox No. 2 (Rorer), Maalox Plus (Rorer), Maalox TC (Rorer), Magna Gel (No. American), Magnatril (Lannett), Mylanta (Stuart), Mylanta II (Stuart), Nephrox (Fleming), Noralac (No. American), Nutrajel (Cenci), Silain-Gel (Robins), Simeco (Wyeth), Syntrogel (Reed and Carrick), Tempo (Richardson-Vicks), Tralmag (O'Neal, Jones, and Feldman), Trimagel (Columbia Medical), Trisogel (Lilly), WinGel (Winthrop)

1b. Aluscop (O'Neal)

1c. Basaljel (Wyeth)

1d. Magnesia and Alumina Oral Suspension (Philips Roxane), Nutramag (Cenci).

1e. Noralac (No. American)

1f. Riopan (Ayerst), Riopan Plus (Ayerst)

1g. Robalate (Robins), Tralmag (O'Neal, Jones, and Feldman)

1h. Roloids (Warner-Lambert)

2a. Arthritis Pain Formula (Whitehall), Ascriptin (Rorer), Ascriptin A/D (Rorer), B-A (O'Neal, Jones, and Feldman), Cama (Dorsey), Cope (Glenbrook), Pabrin (Dorsey), Vanquish Caplet (Glenbrook)

2b. Arthritis Strength Bufferin (Bristol-Myers), Bufferin (Bristol-Myers)

3a. Amogel (No. American), Bislad (Central), Diabismul (O'Neal, Jones, and Feldman), Dia-eze (Central), Donnagel-PG (Robins), Donnagel (Robins), Kaodene Non-Narcotic (Pfeiffer), Kaodene with Paregoric (Pfeiffer), Kaolin Pectin Suspension (Philips Roxane), Kaopectate (Upjohn), Kaopectate Concentrate (Upjohn), Parepectolin (Rorer), Pargel (Parke-Davis), Pektamalt (Warren-Teed)

3b. Pabisol with Paregoric (Rexall)

3c. Quintese (Lilly), Rheaban (Pfizer)

4a. Carafate (Marion Labs)

n.a. = not available

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Human albumin solutions and other biological products intended for human use may contain aluminum because aluminum compounds are used in their manufacture or as a result of contamination. In albumin products, aluminum is generally introduced as a contaminant from filters, filter aides, buffer solutions, anticoagulants, as well as the container itself. Aluminum levels in a 5% pooled human albumin solution was 0.507 µg/mL, (Progar et al. 1996).

5.5 GENERAL POPULATION AND OCCUPATIONAL EXPOSURE

Exposure to aluminum is inevitable due to its natural abundance in the earth's crust and its many uses. The intake of aluminum is chiefly oral, and the major sources for human exposure to aluminum are drinking water, residues in foods, cooking utensils, food and beverage packaging, antacid formulations, and antiperspirant formulations (Marquis 1989). Aluminum is present in the human diet, in amounts varying from relatively low concentrations in animal products to relatively high concentrations in some processed foods. However, the gastrointestinal absorption of aluminum is low (<0.1%) and renal elimination is very effective in removing aluminum in healthy individuals (Muller et al. 1993b).

Aluminum is inhaled from air primarily as aluminosilicates associated with airborne dust particles (Koo and Kaplan 1988). Since a large aqueous concentration of aluminum (i.e., >100 mg/L) can only occur when the pH is < 5 (Sorenson et al. 1974), the levels of aluminum in most natural waters (pH>6) are not expected to be of significant concern to human health. Miller et al. (1984) reported that the median aluminum levels in finished drinking water throughout the United States varied from 0.026 mg/L to 0.161 mg/L (ppm). More recently, Schenck et al. (1989) reported concentrations of aluminum in finished drinking water in various regions of the United States were highly variable, ranging from undetectable to 1.029 mg/L (ppm). The median and mean aluminum concentrations in finished drinking water from 384 Norwegian waterworks sampled on four occasions (autumn 1982, winter, spring, and summer 1983) were 0.06 and 0.11 mg/L (ppm), respectively with a range of < 0.04-4.1 mg/L (Flaten 1991). The median aluminum concentrations in drinking water from 346 surface water and 35 groundwater sources were 0.06 and 0.02 mg/L, respectively. A correlation between aluminum and SO_4^{2-} , Mn, and pH were ascribed to the effects of acid precipitation.

Aluminum is present naturally in tea and some vegetables. Aluminum is introduced into grain products and processed cheese from aluminum-containing food additives. These products are used as acidifiers, buffers, leavening agents, emulsifiers, stabilizers, thickeners, and anticaking agents. For example,

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sodium aluminum sulfate is found in baking powder and sodium silicates are used as anticaking agents in salt and other dry and powdered ingredients. In a report on FDA's Total Diet Study, the foods highest in aluminum were those suspected of containing aluminum additives (e.g., processed cheese, grain products, and grain-based desserts) (Pennington 1987). Measured-daily dietary intakes of aluminum range from 2 to 14 $\mu\text{g}/\text{day}$. The major contributors to aluminum in the diet are grain products (24-49%), dairy products (17-36%), desserts (9-26%), and beverages (5-10%) (Pennington 1987). FDA revised their Total Diet Study in 1991 to reflect current food consumption patterns and to include additional sex-age groups (Pennington and Schoen 1995). Dietary intakes ranged from 0.7 mg/day for infants to 11.5 mg/day for 14-16-year-old males. The aluminum intake of adult males ranged from 8 to 9 mg/day and that for adult females was about 7 mg/day. Dietary intakes for 2-year-old, 6-year-old, and 10-year-old children were 4.6, 6.5, and 6.8 mg/day, respectively. Aluminum intakes per kilogram of body weight were 0.10 mg/kg for infants, 0.35 mg/kg for 2-year-old children, and 0.30 mg/kg for 10-year-old children. The other sex age groups had aluminum intakes of 0.10 to 0.15 mg/kg, except for 14-16-year-old males which was 0.18 mg/kg. More recently, Greger (1992) estimated that most adults consume from 1 to 10 mg aluminum per day from natural sources.

Cooking in aluminum containers often results in statistically significant, but not biologically important, increases in the aluminum content of some foods. In one study, increases in the aluminum content of foods after contact with aluminum utensils were less than 1 mg/kg for 47% of the food examined and less than 10 mg/kg for 85% of the food examined (Pennington and Schoen 1995). However, intake of aluminum from foods containing food additives varies greatly (0-95 mg aluminum/day) among residents of North America, depending on the amount of processed foods consumed. The median intake of aluminum for adults was estimated to be 24 mg/day (Greger 1992). In an Italian study in which samples of daily diets were collected and analyzed, the intake of aluminum ranged from 2.5 to 6.3 mg/day (Gramiccioni et al. 1996). The migration of aluminum from cookware into food was reported to be relatively low. The results of dietary studies indicate that exposure to aluminum through food is low and the total dietary intake of aluminum is correlated with the total food intake. The migration of aluminum from cookware into food will increase with the acidity of the food and the duration of exposure. In a worst-case example, red current juice was prepared by boiling berries for 3 hours in an aluminum or in a stainless steel pot (Valkonen and Aitio 1997). The aluminum concentration of the juice prepared in the aluminum and stainless steel pots were 89.1 mg/L and 1.83 mg/L, respectively. The intake of aluminum in foods is low compared with the amount of aluminum consumed when taking aluminum-containing medication, such as antacids, buffered aspirins, antidiarrheal agents, and certain anti-ulcer drugs at their

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recommended dosages (Lione 1983, 1985a; Pennington and Schoen 1995). Antacids and buffered aspirin, which are often taken in multiple daily doses for prolonged periods, contain 4-562 mg/kg (ppm) of aluminum (Lione 1983; Schenck et al. 1989; Shore and Wyatt 1983). For example, according to Pennington and Schoen (1995), antacids may contain 50 mg of aluminum per tablet and buffered aspirin may contain 10-20 mg of aluminum per tablet. Another source lists the $\text{Al}(\text{OH})_3$ content of several popular antacid preparations (tablet or 5 mL liquid), which range from 400 to 600 mg (140-210 mg of aluminum) (Harman and Limbird 1996).

Reports available on normal dietary levels of aluminum suggest that approximately 20 mg/day may be an acceptable representation (Lione 1983; Underwood 1977). More recently, Greger (1992) reported a median concentration of 24 mg/day, which is comparable. Lione (1985a) estimated that from 126 to 728 mg and 840 to 5,000 mg were possible daily doses of aluminum consumed in buffered aspirins and antacids products, respectively. These doses are from 6 to almost 40 times and 42-250 times greater, respectively, than aluminum doses obtained from consumption of food. When large oral loads of aluminum (1,000-4,000 mg/day) in the form of antacids are ingested, some of this excess aluminum is absorbed, usually less than 1% of the intake amount in healthy individuals (Gorsky et al. 1979, Kaehny et al. 1977; Reiber et al. 1995).

In recent years, a numbers of investigators have become concerned about the aluminum content of infant formulae (Koo et al. 1988; Simmer et al. 1990; Weintraub et al. 1986). The aluminum content of human breast milk or cows' milk is very low ($< 0.05 \mu\text{g}/\text{mL}$ [ppm]) (Koo et al. 1988; Simmer et al. 1990; Weintraub et al. 1986). Dabeka and McKenzie (1990) reported that ready-to-use milk-based and soybased formulae contained 0.01-0.36 and 0.40-6.4 $\mu\text{g}/\text{g}$, respectively. Thus, 1-3-month-old infants consuming certain soy-based formulae could ingest as much as 2.1 mg aluminum a day. This is compared to infants fed human breast milk or cows' milk who would consume only 3 μg aluminum a day. Infants fed the soy-based infant formulae would thereby ingest 700 times more aluminum than infants fed human breast milk or cows' milk (Dabeka and McKenzie 1990; Greger 1992).

As discussed in Section 5.4.2, the median concentration of aluminum in drinking water not receiving coagulation treatment and that receiving coagulation treatment is 0.043 mg/L and 0.112 mg/L, respectively. If the total dose of aluminum obtained from water is calculated based on an estimated consumption of 2 L/day, the amount of aluminum ingested would respectively be 0.08 and 0.224 milligrams per day or roughly 1% of the 7-9 milligrams per day for adults from dietary sources.

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Although the intake of aluminum is chiefly through ingestion of food and drinking water, aluminum is also drawn into the lungs from breathing atmospheric dust (Browning 1969). As discussed in Section 5.4.1, background levels of aluminum in the atmosphere generally range from 0.005 to a maximum of 0.18 ng/m³ in the United States (Hoffman et al. 1969; Sorenson et al. 1974). If the inhalation rate is taken to be 20 m³/day, then the total amount of aluminum obtained from inhalation of 0.18 ng/m³ would be 3.6 nanograms per day, suggesting that ambient air is not normally a major exposure pathway for aluminum. This is negligible compared with the estimated dietary intake for adults of 7-9 milligrams per day. However, the aluminum content of air in urban and industrial areas has been reported to be considerable higher, ranging from 0.4 to 10 ng/m³ (Cooper et al. 1979; Dzubay 1980; Kowalczyk et al. 1982; Lewis and Macias 1980; Moyers et al. 1977; Ondov et al. 1982; Pillay and Thomas 1971; Sorenson et al. 1974; Stevens et al. 1978). If the inhalation rate is taken to be 20 m³/day, then the total amount of aluminum inhaled would range from 8 to 200 nanograms per day, which is still negligible compared with the aluminum intake from dietary sources. Dusts arising from soil, especially in industrial or agricultural areas (Eisenreich 1980), and from the metal surfaces of air conditioners can contain large amounts of aluminum (Crapper-McLachlan 1989), resulting in high localized concentrations and, subsequently, in higher exposures. Typically, however, for the general population, inhalation is likely to be less important as an exposure pathway than is dietary exposure to aluminum but may represent a source of greater exposure in some urban environments.

Because of inherent problems with sensitivity and contamination, levels of aluminum in body tissues are difficult to measure and levels found can be method-dependent (Schenck et al. 1989). Aluminum levels reported in studies prior to 1980 are often much higher than those reported in more recent studies. Normal values of aluminum in whole blood have been reported to range from 0.14 to 6.24 mg/L (ppm), and in plasma from 0.13 to 0.16 mg/L (ppm) (Sorenson et al. 1974). Normal values in serum have been reported at 1.46 and 0.24 mg/L (ppm), using neutron activation and atomic absorption analysis, respectively (Berlyne et al. 1970). A normal value of 0.037 mg/L (ppm) for serum using flameless atomic absorption analysis has also been reported (Fuchs et al. 1974). Drablos et al. (1992) analyzed aluminum serum levels in 230 nonexposed workers (controls) and reported a mean aluminum serum level of 0.005±0.002 mg/L (ppm). Research has shown that the levels of aluminum in the serum in the general population do not exceed 0.01 mg/L (ppm) (Cornelis 1982). Nieboer et al. (1995) reviewed 34 studies on aluminum levels in serum or plasma, and also reported that aluminum serum levels in the general population were typically < 0.01 mg/L (ppm).

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Aluminum concentrations in the urine can serve as an indicator of increased exposure to aluminum because a large proportion of ingested aluminum passes quickly through the body. The normal levels reported in some older studies of aluminum range from 0.05 to 1 mg/L (ppm) in the urine (Kehoe et al. 1940; Tipton et al. 1966). Drablos et al. (1992) analyzed aluminumurine levels in 230 nonexposed workers (controls) and reported a mean aluminum urine level of 0.005 ± 0.003 mg/L (ppm) (range, 0.001-0.037 mg/L). Nieboer et al. (1995) reviewed 8 studies on aluminumlevels in urine and reported that aluminum urine levels in healthy individuals typically ranged from 0.0027 to 0.0081 mg/L (ppm). In a recent Finnish study of aluminum in urine from 3,212 occupationally exposed workers, mostly aluminum welders, between 1993 and 1996, the average annual urinary aluminum level was 1.4 $\mu\text{mol/L}$ (0.038 mg/L) and the range was 1.08-2.04 $\mu\text{mol/L}$ (0.029-0.055 mg/L) (Valkonen and Aitio 1997). The samples, collected as part of a routine occupational health program were collected after the weekend as a morning specimen. The mean urinary aluminum concentration in 44 nonexposed persons, who did not use antacid preparations, was 0.33 $\mu\text{mol/L}$ (0.0089 mg/L), and the range and standard deviation were 0.07-0.82 $\mu\text{mol/L}$ (0.002-0.022 mg/L) and 0.18 $\mu\text{mol/L}$ (0.0022 mg/L), respectively. The mean serum aluminum concentration of 21 of these nonexposed individuals was 0.06 $\mu\text{mol/L}$ (0.0016 mg/L), and the range and standard deviation were 0.02-0.13 $\mu\text{mol/L}$ (0.0005-0.0035 mg/L) and 0.03 $\mu\text{mol/L}$ (0.0008 mg/L), respectively. The mean serumlevel on the nonexposed people was much lower than the mean serum level reported by Drablos et al. (1992). Gitelman et al. (1995) investigated the relationship between the concentration of aluminum in serum and urine and occupational exposure to airborne aluminum in a large number of workers in the aluminum industry (15 plants). Occupational exposure was estimated from aluminum measurements of total and respirable ($< 10 \mu\text{m}$) particulate matter in air. The study showed that workers with occupational exposure to airborne aluminum had statistically significant increases in urinary aluminum/creatinine ratios over controls; however, changes in serum aluminum were borderline. Similarly, in an investigation of workers at an open bauxite mine in Surinam, serum aluminum levels of 24 men working in the mine for an average of 24 years were low and not statistically different from controls (de Kom et al. 1997).

Recent measurements of aluminum concentrations in human tissues for estimation of exposures are primarily limited to bone and brain tissues (Nieboer et al. 1995). Background levels of aluminum in bone are in the order of 1-3 $\mu\text{g/g}$ (ppm, dry weight). These authors also reported that background aluminum levels in brain tissues (primarily grey matter) of healthy individuals typically ranges from 1 to 3 $\mu\text{g/g}$ (ppm, dry weight) or 0.5 $\mu\text{g/g}$ (wet weight).

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Occupational exposure to aluminum occurs not only in the refining of the primary metal, but also in secondary industries that use aluminum products (e.g., aircraft, automotive, and metal products), and aluminum welding (Nieboer et al. 1995). Three major steps are involved in primary aluminum production. Aluminum is first extracted with caustic soda from bauxite ore, precipitated as aluminum hydroxide, and subsequently converted to aluminum oxide in a calcination process. In the second step, the oxide is dissolved in molten cryolite (Na_3AlF_6) and electrolyzed to yield the pure molten metal. The electrolytic cells are called pots and the work area is called the potroom. Casting is the final step in the process where molten aluminum is poured into ingots in the foundry. Exposure is primarily to aluminum hydroxide and oxide in the initial extraction and purification process, to aluminum oxide and aluminum fluoride in the potroom (as well as to tar-pitch volatiles including PAHs), and to partially oxidized aluminum metal fumes in the foundry (Drablos et al. 1992; IARC 1984; Nieboer et al. 1995). Drablos et al. (1992) studied aluminum concentrations in workers at an aluminum fluoride plant. Mean aluminum levels in urine were 0.011 ± 0.007 mg/L (range, 0.002-0.046 mg/L) for 15 plant workers, 0.032 ± 0.023 mg/L (range, 0.006-0.136 mg/L) for 7 foundry workers, and 0.054 ± 0.063 mg/L (range, 0.005-0.492 mg/L) for 12 potroom workers as compared to 0.005 ± 0.003 mg/L (range, 0.001-0.037 mg/L) for 230 unexposed controls.

Most of the studies of occupational exposure (aluminum refining and metal industry workers) to aluminum have dealt with inhalation of aluminum-containing dust particles. Rarely is a worker exposed solely to aluminum-containing dust; however, rather exposure to mixtures of aluminum with fine respirable particles or other toxic chemicals is more prevalent. For example, it had been observed that the incidence of bladder cancer was unusually high among aluminum reduction workers. An epidemiological study showed that volatile PAHs in coal tar pitch, however, were the actual causative agents (Theriault et al. 1984a). Synergism among metal dusts, fine particles, toxic chemicals including PAHs, and cigarette smoke is a highly plausible cause of skin irritation and cancers appearing in workers for many industrial processes involving aluminum.

The most recent National Occupational Exposure Study (NOES) conducted by NIOSH from 1981 to 1983, estimated the number of workers potentially exposed to aluminum and aluminum compounds (NIOSH 1991). Results of this survey are summarized in Table 5-8. The NOES was based on observational field surveys of 4,490 facilities and was designed as a nationwide survey based on a statistical sample of virtually all workplace environments, except mining and agriculture, in the United States where eight or more persons are employed and only provides estimates of the numbers of workers

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Table 5-8. Estimated Number of Workers Potentially Exposed to Aluminum and Its Compounds in the Workplace

Aluminum compound	Number of potentially exposed workers
Aluminum - pure	31,369
Aluminum dust	1,833
Aluminum - unknown	1,033,235
Aluminum oxide	1,345,659
Aluminum oxide, powder	172,756
Aluminum hydroxide	325,788
Aluminum hydroxide, gel	37,772
Dried aluminum hydroxide gel	7,006
Aluminum chloride	49,913
Aluminum chloride hydroxide	1,579
Aluminum sulfate	212,239
Aluminum sulfate, liquid	23,354
Aluminum sulfate, powder	1,496
Aluminum nitrate	34,929
Aluminum phosphide	622
Aluminum phosphate	19,526
Aluminum phosphate, gel	4,228
Aluminum fluoride	175
Aluminum, calcined	27,670

Source: National Occupational Exposure Study (NOES) NIOSH 1991, 1992

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potentially exposed to chemicals in the workplace (NIOSH 1988). It does not contain information on the frequency, concentration, or duration of occupational exposure to any of the chemicals listed. The industries with the largest numbers of workers potentially exposed to aluminum and aluminum compounds include: plumbing, heating, and air conditioning; masonry and other stonework; electrical work; machinery except electrical; certified air transportation equipment; electrical components; fabricated wire products; general medical and surgical hospitals; industrial buildings and warehouses, and special dies, tools, jigs, and fixtures.

5.6 EXPOSURES OF CHILDREN

This section focuses on exposures from conception to maturity at 18 years in humans and briefly considers potential pre-conception exposure to germ cells. Differences from adults in susceptibility to hazardous substances are discussed in Section 2.6.

Children are not small adults. A child's exposure may differ from an adult's exposure in many ways. Children drink more fluids, eat more food, and breathe more air per kilogram of body weight, and have a larger skin surface in proportion to their body volume. A child's diet often differs from that of adults. The developing human's source of nutrition changes with age: from placental nourishment to breast milk or formula to the diet of older children who eat more of certain types of foods than adults. A child's behavior and lifestyle also influence exposure. Children crawl on the floor; they put things in their mouths; they may ingest inappropriate things such as dirt or paint chips; they spend more time outdoors. Children also are closer to the ground, and they do not have the judgement of adults in avoiding hazards (NRC 1993).

As with adults, exposures of children to aluminum from breathing air, drinking water, and eating food is low. However, children are much more likely to ingest dirt, which contains high amounts of aluminum than adults. They are likely to ingest dirt from their unwashed hands or when playing with contaminated soils. In addition, children living in proximity to hazardous waste sites may be exposed to aluminum via ingestion of aluminum contained in soil, or via inhalation of aluminum from soil that is entrained in air. While aluminum contained in dirt may be in many forms, some of these forms may be embedded in minerals not bioavailable even in the acid environment of the stomach. Aluminum found at hazardous waste sites may be in a more labile form than that found in ordinary soil.

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When FDA revised their Total Diet Study in 1991, several sex-age groups relating to children were included (Pennington and Schoen 1995). Average dietary intakes of aluminum in children are shown in Table 5-9. Dietary intakes of aluminum for children ranged from 0.7 mg/day for infants to 11.5 mg/day for 14-16-year-old males. Aluminum intakes per kilogram of body weight for children ranged from 0.10 mg/kg for infants to 0.35 mg/kg for 2-year-old children. The major sources of aluminum in food by age-sex group is shown in Table 5-10. Processed foods containing aluminum additives such as processed cheese and grain-based products constitute the foods with the largest quantities of aluminum and the largest components of the dietary intake of children. Soy-based formula may contain high quantities of aluminum and infants on such formula would have much higher dietary intakes of aluminum than other infants. A comparison of aluminum concentrations in breast milk, humanized infant formulas, and special purposed infant formulas appears in Table 5-5.

Aluminum levels have also been reported for human breast milk. The median aluminum level in breast milk collected from 12 Canadian women was reported to be 14 $\mu\text{g/L}$ (ppb, range < 5-45 $\mu\text{g/L}$) (Koo et al. 1988). In an Australian study, Weintraub et al. (1986) reported human breast milk concentrations of 30 $\mu\text{g/L}$ (ppb) in nursing mothers. More recently, Simmer et al. (1990) reported a mean aluminum concentration of 49 $\mu\text{g/L}$ (ppb) in breast milk collected from Australian women. Hawkins et al. (1994) reported breast milk aluminum concentrations of 9.2 $\mu\text{g/L}$ (ppb) (95% confidence interval from 5.6 to 12.7 $\mu\text{g/L}$) collected from 15 nursing mothers in the United Kingdom. The aluminum content of human milk from 42 nursing Croatian women in the winter of 1992-1993 ranged from 4 to 2,670 $\mu\text{g/L}$ (ppb) with a mean of 380 $\mu\text{g/L}$ (ppb) (Mandic et al. 1995). While some differences in aluminum content of milk was found depending the participant's age, number of deliveries, postpartum days, weight gain during pregnancy, refugee status, and smoking status, correlations with these factors were not statistically significant. The investigators were unable to explain the high values obtained for aluminum in the milk of the Croatian women, especially since there was no data on aluminum in Croatian foodstuffs. Since their measurements using standard reference serum were acceptable, contamination in the analytical procedure was ruled out. While steps were taken to avoid contamination in the collection process, no controls to gauge the effectiveness of these steps were reported.

As with adults, aluminum intake from aluminum-containing medication, such as antacids, buffered aspirins, and antidiarrheal agents would overwhelm ordinary dietary intakes (Pennington and Schoen 1995). Children may also be exposed to aluminum from vaccinations, parenteral feeding of premature infants, dialysis fluids, and treatment for hyperphosphatemia.

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Table 5-9. Dietary Intakes of Aluminum in Children

Age-sex group	Aluminum Intake	
	(mg/day)	(mg/kg)
6–11-month-old infants	0.7	0.10
2-year-old children	4.6	0.35
6-year-old children	6.5	0.30
10-year-old children	6.8	0.11
14–16-year-old females	7.7	0.15
14–16-year-old males	11.5	0.18

Source: Pennington and Schoen 1995.

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Table 5-10. Major Sources of Aluminum in Food by Age-Sex Group

Foods by age-sex group (Al/day)	Aluminum/day	
	mg	% of total intake
6-11-month-old infants (0.7 mg)		
Soy-based formula	0.161	23.0
American processed cheese	0.122	17.4
Yellow cake with icing	0.088	12.6
Green beans, strained	0.038	5.4
Pancakes	0.029	4.1
Total	0.438	62.6
2-year-old children (4.6 mg)		
Cornbread	1.580	34.3
American processed cheese	1.037	22.5
Yellow cake with icing	0.384	8.3
Fish sticks	0.173	5.4
Pancakes	0.113	2.5
Tortillas	0.093	2.0
Muffins	0.093	2.0
Fruit drink from powder	0.079	1.7
Taco/tostada	0.071	1.5
Tea	0.061	1.3
Total	3.684	80.1
6-year-old children (6.5 mg)		
American processed cheese	1.382	21.3
Yellow cake with icing	1.091	16.8
Pancakes	0.752	11.6
Fish sticks	0.529	8.1
Cornbread	0.450	6.9
Tortillas	0.297	4.6
Taco/tostada	0.209	3.2
Muffins	0.202	3.1
Hamburger	0.104	1.6
Fruit drink from powder	0.105	1.6
Total	5.121	78.8
10-year-old children (6.8 mg)		
American processed cheese	1.498	22.0
Cornbread	1.105	16.3
Pancakes	0.858	12.6
Tortillas	0.344	5.1
Yellow cake with icing	0.350	5.1
Fish sticks	0.280	4.1
Taco/tostada	0.259	3.8
Muffins	0.207	3.0
Chocolate cake with icing	0.141	2.1

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Table 5-10. Major Sources of Aluminum in Food by Age-Sex Group (continued)

Foods by age-sex group (Al/day)	Aluminum/day	
	mg	% of total intake
Chocolate snack cake	0.144	2.1
Total	5.186	76.3
14-16-year-old females (7.7 mg)		
American processed cheese	2.139	27.8
Yellow cake with icing	0.906	11.8
Cornbread	0.781	10.1
Taco/tostada	0.682	8.9
Pancakes	0.668	8.7
Tortillas	0.325	4.2
Muffins	0.219	2.8
Cheeseburger	0.183	2.4
Tea	0.159	2.1
Fish sticks	0.125	1.6
Total	6.187	80.4
14-16-year-old males (11.5 mg)		
Cornbread	4.209	36.6
American processed cheese	1.978	17.2
Pancakes	1.038	9.0
Yellow cake with icing	0.925	8.0
Taco/tostada	0.398	3.5
Tortillas	0.398	3.5
Cheeseburger	0.310	2.7
Tea	0.225	2.0
Hamburger	0.211	1.8
Fish sticks	0.170	1.5
Total	9.862	85.8

Source: Pennington and Schoen 1995

5.7 POPULATIONS WITH POTENTIALLY HIGH EXPOSURES

In addition to individuals who are occupationally exposed to aluminum (see Section 5.5), there are several groups within the general population that have potentially higher exposures (higher than background) than the general population. These populations include members of the general population living in the vicinity of industrial emission sources and hazardous waste sites, individuals with chronic kidney failure requiring long-term hemodialysis treatment, infants fed a formula diet containing high levels of aluminum and individuals consuming large quantities of antacid formulations for gastric disorders, anti-ulcerative medications, buffered analgesics for arthritis, or antidiarrheal medications. Furthermore, the elderly are at risk because of multiple chronic diseases including ulcers and other gastrointestinal diseases, rheumatoid arthritis, and renal disorders. Aluminum has been detected in virtually all food products (especially plant-derived and processed foods), ambient air, drinking water, and soils. Substantially higher concentrations of aluminum have been detected in localized areas around some industrial and hazardous waste disposal sites. However, exposure to higher levels of aluminum may not be hazardous if the exposed individual has normal renal function (see Section 2.5).

Individuals living or working in proximity to aluminum production facilities may be exposed to higher concentrations of aluminum in the ambient air than members of the general population. Aluminum has been detected in air samples collected at 9 of the 427 NPL hazardous waste sites where it has been detected in some environmental media (HazDat 1996). In addition individuals living in proximity to hazardous waste sites may be exposed to aluminum via ingestion of aluminum contained in soil from their unwashed hands when working or playing with contaminated soils and sediments. Children in particular are likely to ingest dirt from their unwashed hands, or inhale resuspended dust during near-ground activities. Aluminum has been detected in soil and sediment samples at 203 and 151 of 427 NPL hazardous waste sites, respectively, where it has been detected in some environmental media (HazDat 1996). If residential wells are the primary source of drinking water, this may also pose a risk to human health via consumption of contaminated drinking water. Aluminum has been detected in groundwater at 336 of 427 NPL hazardous waste sites where it has been detected in some environmental media (HazDat 1996).

Individuals with chronic renal failure requiring long-term hemodialysis treatment are another group within the general population that may be exposed to greater than background levels of aluminum (Alfrey 1987; Lione 1985a; Muller et al. 1993b). Aluminum levels in virtually every body tissue are

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significantly higher in this group of patients if aluminum is present in the dialysate (Alfrey et al. 1980; Cooke and Gould 1991). In addition, Ward (1991) reported increased serum aluminum concentrations in hemodialysis patients who were prescribed effervescent analgesic tablets which increased gastrointestinal absorption mediated by the acidity of citric acid that increased aluminum solubility and therefore availability of aluminum for uptake.

The oral intake of aluminum tends to be higher for children than for adults (Greger 1992). Calculations based on the FDA's Total Diet Study suggest that 2-year-olds (13 kg body weight) consumed almost 3 times as much aluminum per kg body weight as adult males (75 kg body weight) or adult females (60 kg body weight), respectively (0.48 vs. 0.18 and 0.15 mg aluminum/kg body weight) (Greger 1992). Infants fed milk-based or soy-based infant formulae can be exposed to considerably higher concentrations of aluminum than infants fed breast milk or cow's milk (see Section 5.4.4). Within this group, the infants believed to be most at risk would be preterm infants with impaired renal function because they would be less able to excrete the absorbed aluminum (Bishop 1992; Greger 1992; Koo et al. 1988, 1992; Weintraub et al. 1986).

As discussed in Section 5.4.4, individuals consuming large quantities of antacid formulations, anti-ulcerative medications, buffered analgesics, or antidiarrheal medications are exposed to higher than background doses of aluminum in their diet. Lione (1985a) estimated that from 126 to 728 mg and 840 to 5,000 mg were possible daily doses of aluminum consumed in buffered aspirins for rheumatoid arthritis and antacid products, respectively. These doses are from 6 to 40 times and 42 to 250 times greater, respectively, than aluminum doses obtained from consumption of foods (20-24 mg/day).

5.8 ADEQUACY OF THE DATABASE

Section 104(i)(5) of CERCLA, as amended, directs the Administrator of ATSDR (in consultation with the Administrator of EPA and agencies and programs of the Public Health Service) to assess whether adequate information on the health effects of aluminum is available. Where adequate information is not available, ATSDR, in conjunction with the NTP, is required to assure the initiation of a program of research designed to determine the health effects (and techniques for developing methods to determine such health effects) of aluminum.

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The following categories of possible data needs have been identified by a joint team of scientists from ATSDR, NTP, and EPA. They are defined as substance-specific informational needs that if met would reduce the uncertainties of human health assessment. This definition should not be interpreted to mean that all data needs discussed in this section must be filled. In the future, the identified data needs will be evaluated and prioritized, and a substance-specific research agenda will be proposed.

5.8.1 Identification of Data Needs

Physical and Chemical Properties. The physical and chemical properties of aluminum and various aluminum-containing compounds are sufficiently well defined to allow an assessment of the environmental fate of these compounds (Budavari et al. 1989; HSDB 1995; Lewis 1993; Sax and Lewis 1989; Weast et al. 1989; Weiss 1986).

Production, Import/Export, Use, Release, and Disposal. Because aluminum compounds occur naturally (Browning 1969; Dinman 1983; IARC 1984; NRC 1982) and are widely used in industry, in the manufacture of household products, and in processing, packaging, and preserving food (Browning 1969; Budavari et al. 1989; Hawley 1977; Sax and Lewis 1987; Stokinger 1981; Venugopal and Lucky 1978), the potential for human exposure to these compounds through ingestion of food and water and inhalation of airborne particulates is substantial. Recent information on production volumes is available and it appears that, while primary production of aluminum has decreased from 1991 to 1996, secondary recovery of aluminum (recycling) increased during this same period (USGS 1996, 1997a). The United States relies on imports for some of its consumption needs; however, imports declined slightly in 1995, reversing an increasing trend that began in 1992 (USGS 1996, 1997a). Exports have remained relatively constant from 1991 through 1996 (USGS 1996, 1997a). Consumption data for aluminum used in areas impacting on exposure such as food additives are not available. Information on disposal of aluminum compounds is limited. Additional information on disposal would be useful in assessing the potential for the release of and exposure to aluminum compounds.

According to the Emergency Planning and Community Right-to-Know Act of 1986, 42 U.S.C. Section 11023, industries are required to submit chemical release and off-site transfer information to the EPA. The Toxics Release Inventory (TRI), presently contains this information for 1996. This database will be updated yearly and should provide a list of industrial production facilities and emissions.

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Environmental Fate. Aluminum partitions to air, water, soil, and plant material. Its partitioning to various media is determined by the physical and chemical properties of the aluminum compound and the characteristics of the environmental matrix that affects its solubility (Brusewitz 1984; Dahlgren and Ugolini 1989; Filipek et al. 1987; Goenaga and Williams 1988; James and Riha 1989; Litaor 1987; Mulder et al. 1989; Wangen and Jones 1984). Aluminum is transported through the atmosphere primarily as a constituent of soil and other particulate matter (Eisenreich 1980). Transformations are not expected to occur during transport of aluminum through the atmosphere. Aluminum partitions between solid and liquid phases by reacting and complexing with water molecules, anionic compounds, and negatively charged functional groups on humic materials and clay (Bodek et al. 1988). Information on the environmental fate of aluminum is sufficient to permit a general understanding of transport and transformation in all environmental media. No additional information is needed at this time.

Bioavailability from Environmental Media. Aluminum compounds are deposited in the lungs following inhalation (Christie et al. 1963; Steinhagen et al. 1978; Stone et al. 1979; Thomson et al. 1986) and are poorly absorbed following ingestion (Cranmer et al. 1986). Very limited information is available regarding absorption following dermal contact; however, this pathway of exposure is not expected to be significant. Additional information on absorption following ingestion of soils contaminated with aluminum compounds and dermal contact would be useful in assessing bioavailability following exposure via these routes.

Food Chain Bioaccumulation. Little information is available on the uptake of aluminum into food crops. Uptake into root crops is of particular importance, since many plant species concentrate aluminum in their roots (Baes et al. 1984; Kabata-Pendias and Pendias 1984; Vogt et al. 1987). The limited information available on bioconcentration in animals appears to indicate that aluminum is not significantly taken up by livestock (Baes et al. 1984). The fact that in studies dealing with aluminum in food, aluminum is generally present in low concentrations in fruit, vegetables, and meat products that do not contain aluminum additives or have other contact with aluminum (e.g., cooked in aluminum pots) (Pennington and Schoen 1995), would support a conclusion that aluminum does not bioaccumulate in the food chain. Because of its toxicity to many aquatic species, aluminum does not bioconcentrate appreciably in fish and shellfish and therefore it would not be a significant component of the diet of animals that feed upon them (Rosseland et al. 1990). Further studies on the uptake of aluminum by plants, especially those grown on acid soils, would be useful in expanding a somewhat limited database

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and characterizing the importance of food chain bioaccumulation of aluminum as a source of exposure for particular population groups.

Exposure Levels in Environmental Media. There are reliable data to characterize the potential for human exposure from drinking water (Kopp and Kroner 1970; Letterman and Driscoll 1988; Miller et al. 1984; Schenck et al. 1989) and food sources (Brusewitz 1984; Connor and Shacklette 1975; Koo and Kaplan 1988; Lewis 1989; Pennington 1987; Schenck et al. 1989; Sorenson et al. 1974; Weintraub et al. 1986). However, recent (i.e., within 3 years) monitoring data for all media are currently not available. Estimates of human exposure to aluminum from food (Greger 1992; Lione 1983; Pennington 1987; Underwood 1977), drinking water (Kopp and Kroner 1970; Miller et al. 1984; Schenck et al. 1989), and air (Browning 1969; Crapper-McLachlan 1989; Sorenson et al. 1974) are available as are estimates from exposure from antacids, buffered analgesics, antidiarrheal and anti-ulcerative compounds (Lione 1985a; Schenck et al. 1989; Shore and Wyatt 1983). Information on the intake of aluminum from vitamins and other dietary supplements is lacking and would be useful in estimating human exposure. Additional information on the occurrence of aluminum in the atmosphere, surface water, groundwater, and soils surrounding hazardous waste sites would be helpful in updating estimates of human intake.

Reliable monitoring data for the levels of aluminum in contaminated media at hazardous waste sites are needed so that the information obtained on levels of aluminum in the environment can be used in combination with the known body burdens of aluminum to assess the potential risk of adverse health effects in populations living in the vicinity of hazardous waste sites.

Exposure Levels in Humans. Measurements of the aluminum content in human tissues, especially in blood (Berlyne et al. 1970; Cornelis 1982; Drablos et al. 1992; Fuchs et al. 1974; Nieboer et al. 1995; Sorenson et al. 1974), urine (Alessio et al. 1989; Drablos et al. 1992; Kehoe et al. 1940; Nieboer et al. 1995; Tipton et al. 1966), and breast milk (Hawkins et al. 1994; Koo and Kaplan 1988; Simmer et al. 1990; Weintraub et al. 1986), are available. Measurements of aluminum in bone and brain tissue are also available (Nieboer et al. 1995). However, recent (i.e., within 3 years) biological monitoring data, particularly for aluminum in blood and urine, are limited. More recent information would be useful in assessing current exposure levels. Additional biological monitoring data for populations surrounding hazardous waste sites would be useful in helping to better characterize human exposure levels. This information is necessary for assessing the need to conduct health studies on these populations.

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Exposures of Children. Measurements of the aluminum content in tissues, blood, and urine of children who have been exposed to aluminum as well as unexposed children, are not available. This information would be useful in assessing both the normal aluminum content of children and the effect of exposure on aluminum levels in children. This information would also be useful in assessing differences in the effect of aluminum exposure on children to that of adults. While the largest source of aluminum exposure in adults is from aluminum-containing medications and cosmetics, we do not know the amount of such products that may be given to children. We also do not know the intake of available aluminum from soil during childhood activities, or the placental transfer to fetal blood, especially among pregnant women taking antacids as a result of abdominal upsets. Such information would be useful in assessing exposure levels in children.

Data are available on the intake of aluminum in food eaten by children and from their diet (Dabeka and McKenzie 1990; Koo et al. 1988; Pennington and Schoen 1995; Pennington 1987; Simmer et al. 1990; Weintraub et al. 1986). We also know that the aluminum content of human breast milk or cow's milk is very low ($< 0.05 \mu\text{g/mL}$ [ppm]) (Koo et al. 1988; Simmer et al. 1990; Weintraub et al. 1986).

Exposure Registries. No exposure registries for aluminum were located. This substance is not currently one of the compounds for which a subregistry has been established in the National Exposure Registry. The substance will be considered in the future when chemical selection is made for subregistries to be established. The information that is amassed in the National Exposure Registry facilitates the epidemiological research needed to assess adverse health outcomes that may be related to exposure to this substance.

5.8.2 Ongoing Studies

A search of Federal Research in Progress (FEDRIP 1996) identified numerous research studies that are currently being conducted that may fill some of the data needs for aluminum discussed in Section 5.8.1. These studies are summarized in Table 5- 11.

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Table 5-11. Ongoing Studies on Aluminum

Investigator	Affiliation	Research description	Sponsor
Adams J	Auburn University, Auburn, AL	Research on the effects of ecosystems on soil acidity and aluminum toxicity to determine the effects of organic matter derived from forest and agricultural ecosystems on soil pH and aluminum content, determine the quantity and rate of production of low-molecular-weight di- and tri-carboxylic organic acid production in forest and agricultural ecosystems to evaluate aluminum-organic acid complex equilibrium in soils.	U.S. Department of Agriculture
Bondy SC	University of California, Irvine, CA	There is suspicion that aluminum is involved in several neurological diseases associated with aging and there is evidence the potential of iron for enhancing free radical generation in nervous tissue is enhanced by aluminum. It is hoped the results of this project will so reveal.	U.S. Department of Health And Human Services; Public Health Service; National Institute of Health, National Institute of Environmental Health Sciences
Burau R	University of California, Land, Air, and Water Resources Department	Conducting work under the National Atmospheric Deposition Program. As part of this project, acid metal-containing surface waters will be neutralized to determine the degree to which dissolved iron and aluminum can be precipitated and the degree to which these materials can remove other toxic trace elements. The studies will include a characterization of precipitates as well as a determination of the factors which affect the rates of formation of the oxides and oxyhydroxides.	U.S. Department of Agriculture

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Table 5-11. Ongoing Studies on Aluminum (*continued*)

Investigator	Affiliation	Research description	Sponsor
Cavallaro N and Snyder V	University of Puerto Rico, Agronomy and Soils Department, Mayaguez, Puerto Rico	Conducting research on the mobility of aluminum and major nutrient cations in acid ultisols and oxisols. The objective of this study is to evaluate movement and changes in the exchangeable fraction and soluble forms of major cations and anions in a soil profile with different sources and methods of application of materials to reduce soil acidity.	U.S. Department of Agriculture
Etherton B and Cumming J	University of Vermont, Botany Department, Burlington, VT	Conducting a study of membrane transport processes during aluminum exposure in the bean (<i>Phaseolus vulgaris</i>). The objective of this study is to measure net ion fluxes at the root apex during exposure to aluminum and the acquisition of aluminum tolerance, and to determine the role of organic acids in conferring aluminum tolerance in <i>P. vulgaris</i> .	U.S. Department of Agriculture
Golub MS	University of California, Davis, CA	Using mice, the objective of this project is to determine biological actions relevant to toxicological effects, and clarify potential human health risks associated with ingestion of aluminum in food, water, and pharmaceuticals.	U.S. Department of Health and Human Services; Public Health Service; National Institute of Health, National Institute of Environmental Health Sciences
Grunes D and Norvell W	Agricultural Research Service, Ithaca, NY	Work on factors limiting the availability and movement of nutrients in soil. This research will examine factors limiting the availability of nutritionally important elements in soil, movement of these elements to the root-soil interface, and their uptake by plants. Measurements will be made of the form and levels of elements in soil, movement of elements to plant roots, uptake of elements by plant roots, and translocation to above-ground, and edible portions of plants. The elements to be studied include magnesium, calcium, potassium, zinc, phosphorous, nitrogen, iron, manganese, copper, and aluminum.	U.S. Department of Agriculture

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Table 5-11. Ongoing Studies on Aluminum (continued)

Investigator	Affiliation	Research description	Sponsor
Lewis Lovelace JL	Biomed Environ Res In, Albuquerque, NM	Determination of the specific disposition of inhaled toxicants in the olfactory system; assessment of the importance as a human health risk factor of the phenomenon of olfactory transport.	U.S. Department of Health and Human Services; Public Health Service; National Institute of Health, National Institute on Deafness and Other Communication Disorders
Longnecker MP	NIEHS, NIH	Toenail levels may provide a means of measuring exposure for a group of 12 elements that is linked with chronic disease, because toenails reflect exposure over a longer period of time than blood or urine, and are less influenced by contamination.	U.S. Department of Health and Human Services; Public Health Service; National Institute of Health, National Institute of Environmental Health Sciences
Murdoch P	U.S. Geological Survey in Southeastern New York	A study of biogeochemical processes controlling nitrogen cycling and associated hydrogen and aluminum leaching in an undeveloped headwater basin of the Delaware River. Nitric acid is the primary mineral acid causing pH depressions and increases in inorganic aluminum concentrations in streams during storms and snowmelt in the Catskill Mountains of New York. Processes controlling nitrogen movement in forested catchments is poorly understood. An understanding of nitrogen processes and movement in forest soils would allow greater insight into the flowpath of water and the transport of toxic aluminum in watersheds.	Department of the Interior

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Table 5-11. Ongoing Studies on Aluminum (continued)

Investigator	Affiliation	Research description	Sponsor
Norwell W, Grunes D, and Duxbury J	Cornell University, Ithaca, NY	A project on plant availability and geographic distribution of essential and toxic elements. Develop useful maps of nutrient availability and toxic element distribution by using geostatistics and graphical information system techniques to interpret plant and soil composition along with soil genetic information and geological data. Determine effects of aluminum and root exudates on the uptake and translocations of magnesium, calcium, and potassium by aluminum-sensitive and aluminum-tolerant wheat seedlings.	U.S. Department of Agriculture
Robarge W	North Carolina State University, Soil Sciences Department, Raleigh, NC	Research to enhance understanding of how soils can either be a source or can ameliorate various airborne pollutants. As part of this project they will develop a stochastic model of the effect of acidic deposition on the activity of aluminum in soil ecosystems.	U.S. Department of Agriculture
Rufy T	North Carolina State University, Crop Sciences Department Raleigh, NC	Research on the mechanism of aluminum toxicity in plants. This study will determine the extent of aluminum accumulation inside cells at the root apex using microanalytical techniques and will define associated effects on cell division, cell expansion, and cellular accumulation of calcium and magnesium.	U.S. Department of Agriculture
Sakhaee K	University of Texas Southwest Medical Center, Dallas, TX	A project on aluminum absorption and the effect of calcium citrate on aluminum-containing antacids. The objective of this study is to determine whether calcium citrate given together with aluminum-containing antacids would enhance intestinal absorption of aluminum in humans.	National Center for Research Resources

5. POTENTIAL FOR HUMAN EXPOSURE

Table 5-11. Ongoing Studies on Aluminum (continued)

Investigator	Affiliation	Research description	Sponsor
Stilwell D	Connecticut Agricultural Experiment Station, New Haven, CT	Research on the heavy metal content of municipal solid waste. This project will develop methods of analysis and determining the variability of heavy metals and other selected elements in composted municipal solid waste (MSW). Analytical methods will be developed for the determination of several heavy metals, and other selected elements including aluminum, arsenic, boron, calcium, potassium, molybdenum, nitrogen, and phosphorous.	U.S. Department of Agriculture
Zasoski R	University of California, Land, Air, and Water Resources Department, Davis, CA	A study as part of the National Atmospheric Deposition Program which provides the scientific community, resource managers, and policy makers with information on the exposure of both natural and managed ecosystems to biologically important chemical deposition and other stresses resulting from changes in the chemical climate. Acid soils of natural and anthropogenic origins will be characterized for various elements including aluminum and utilized to grow cultivated and native tree species with the objective of determining the composition and character of the rhizosphere associated with these species.	U.S. Department of Agriculture